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Some zoned skarns from granite–marble contacts near Puyvalador, in the Quérigut area, eastern Pyrenees, and their petrogenesis.

(With Plate XIX.)

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Summary. Descriptions, with chemical and mineralogical data, are given of zoned wollastonite–grossular–clinozoisite skarns found at the contact of limestone and granite at Puyvalador, near Quérigut, France. It is shown that the silica and alumina necessary for the formation of the skarn were supplied by the adjacent granite, which in return received lime from the marble replaced by the skarn zones. The modified facies of the granite in contact with the skarn is also described; it shows evidence of migration of silica towards the contact and of accumulation of lime derived from the neighbouring marble.

Similar, but not identical, changes are found at other localities along the Quérigut granite contact with limestone. The features of the Puyvalador granite–limestone contacts are compared and contrasted with those described from some other regions. The contact rocks dealt with here are characterized by the presence of abundant clinozoisite, indicative of relatively low temperatures and abundance of water at the contact in the later stages of evolution of the granite.

THE zoned skarn rocks to be described below were collected from near the southern border of the granite massif of Quérigut–Millas, one of several acid intrusions probably of Carboniferous age cutting the lower and middle Palaeozoic rocks of the eastern Pyrenees.[1] The granite and the contact metamorphic effects induced by it in the surrounding

[1] The general geological structure of the eastern part of the Pyrenees has been briefly described by Raguin (1938) and Abrard (1948); a summary of recent views on the relationships between the main groups of older rocks in the central and eastern Pyrenees is given by de Sitter (1956).

beds were described by Lacroix, in two detailed and now classic memoirs (1898, 1900), which remain the best description of the rocks in the Quérigut area. In this work Lacroix gave a convincing demonstration of the importance of materials added to the sedimentary rocks from the granite magma during the period of metamorphism. The Quérigut area has thus been well known for many years as a result of Lacroix's descriptions of the felspathization of the schists bordering the intrusion, and of the appearance of new minerals, particularly garnet and epidote, in the limestones within the aureole of the granite. As his main conclusion, Lacroix affirmed that large-scale contamination of the granite by the calcareous and interbanded calcareous and siliceous contact rocks led to the production of hornblende-granite, which is observed along parts of the contact of the main intrusion with the calcareous or calcareo-siliceous rocks, and of more basic rocks (diorites, norites, hornblende-peridotites), small bodies of which are observed at several localities. This paper will deal only in passing with the genesis of these intermediate and basic rocks.

The Quérigut–Millas intrusion forms an elongate mass of granite, some 55 km. in length, lying between Ax-les-Thermes and Perpignan (see inset to fig. 1). The accompanying sketch-map (fig. 1), based mainly on the map given by Lacroix (1900), with additions from the detailed map of the French Geological Survey (Quillan Sheet, 1:80 000, 1938), shows the distribution of the main groups of rocks in the western part of the Quérigut–Millas intrusion. These have been fully described by Lacroix, and at present reference will be made only to the limestone and to the interbanded calcareous and siliceous rocks forming much of the enclosed belt of meta-sedimentary rocks near the southern border of the intrusion (the *zone enclavée* of Lacroix). The petrography of the granite and some of its modified facies are referred to in a later section of this paper.

Preliminary laboratory work early in 1954 was carried out on specimens brought from the Quérigut district several years previously by members of the Department of Mineralogy and Petrology, Cambridge University. Later, the writer had the opportunity of visiting the field and examining a number of the contacts. The main localities referred to in the following pages are indicated on the accompanying sketch-map (fig. 1).

The limestones and the calcareo-siliceous rocks: general description.

The unmetamorphosed limestone is a fine-grained, massive rock, light to dark grey in colour. Along the granite contact it is recrystallized,

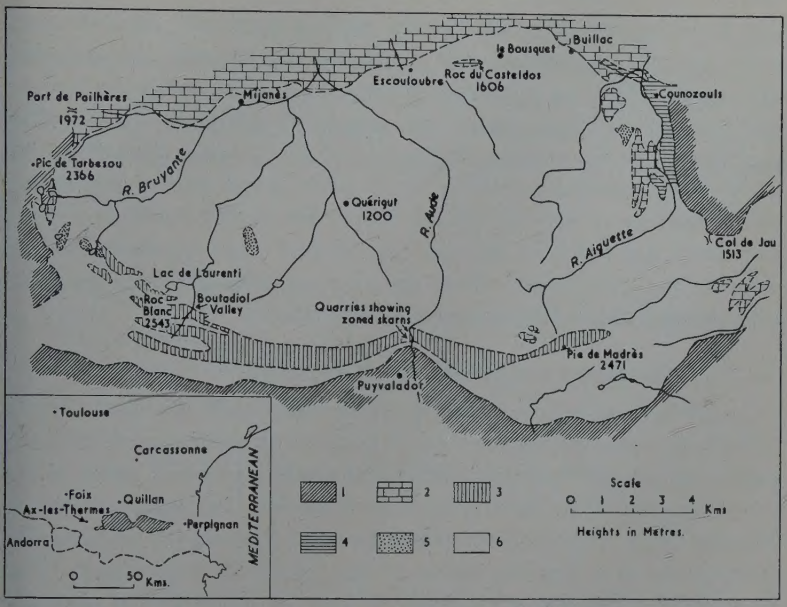

FIG. 1. Sketch-map of the western part of the Quérigut–Millas intrusion. 1. Lower Palaeozoic rocks, mainly argillaceous schists. 2. Middle Palaeozoic, mainly limestone. 3. Interbanded calcareous and siliceous rocks (Devonian). 4. Liassic sediments. 5. Intermediate to basic igneous rocks. 6. Granite, principally porphyritic biotite-granite, but including hornblende-granite near the boundary with calcareous rocks in many places on the southern part of the intrusion. (Inset map shows the general location of the Quérigut-Millas intrusion.)

usually with noticeable increase in grain-size, to a white or light yellowish marble. This is generally hard and compact, though occasionally weathered on the surface to a soft friable crust. In nearly every specimen examined it is composed very largely of calcite, but dolomite also occurs at a few localities. When the limestone comes into contact with the granite a skarn zone is sometimes developed, the skarns associated with dolomitic marble being distinct from those associated with the normal calcite marbles.

Metamorphism of the calcareous rocks interbedded with quartzitic or argillaceous bands has produced a variety of calc-silicate rocks (see Lacroix, 1898, pp. 16–17, 26–28, 39–40; 1900, pp. 7–9, 14–15). These are massive, hard, sometimes flinty rocks, often banded, and generally pinkish or greenish in colour. Their composition, dependent mainly on the relative amounts of limestone and silicate rocks in the original sediments, is very variable; nevertheless, the following four broad groups

may be recognized: grossular-bearing marbles grading to coarse-grained green and pink calc-silicate rocks (including the *grenatites* and *épidotites* of Lacroix), which are sometimes imperfectly banded; calc-silicate rocks carrying potash felspar, which are reasonably common and are often encountered in the vicinity of granite and aplite veins, towards which latter there sometimes appears to be more or less perfect gradation; calc-silicate rocks carrying biotite, only a few of which were recorded; and hornfelses (with or without hornblende) made up essentially of diopside and plagioclase, which are often interbanded with the calc-silicate rocks of the first two groups noted above.

Occurrence and petrography of the zoned skarn rocks.

Zoned skarn rocks have been found at several localities in the Quérigut district where the granite, or veins of granite and aplite, are observed in contact with nearly pure carbonate rock; they may occur either in the limestone proper, or in limestone members of the banded calcareo-siliceous rocks. The finest specimens seen occur among the calcareo-siliceous rocks at a locality about $1\frac{1}{2}$ km. north of Puyvalador. The Puyvalador zoned skarns are found in quarries in the belt of calcareo-siliceous rocks at the point where it meets the Aude river. Fig. 2 shows a plan and sections of the quarries, which open on to a secondary road (D 32, leading from the main road [RN 118] to Quérigut). Here the belt of calcareo-siliceous rocks, bordered on both sides by biotite-rich granite, is exposed along the Aude valley between the road junction and a small stream that enters the river a little over 400 metres to the north. The road section (fig. 2, A and B) shows good exposures of banded calc-silicate rocks with several marble bands and layers of dark diopside–plagioclase hornfels. Veins of granitic rock (up to 3 or 4 metres in thickness) cut the meta-sedimentary rocks at several places, but the contacts are sometimes partly hidden by drift. The large quarry (quarry 2) near the north end of the section is cut into banded epidote- and garnet-rich calc-silicate hornfelses with ribs of marble, and these rocks are seen here in contact with thin veins of granite.

Section C (fig. 2) shows a sketch across the large, now disused quarry (quarry 1) at the road junction. The main rock exposed in it is an almost pure, white marble, which grades near the southern edge of the quarry, before the granite contact is reached, to a fine-grained grey marble with large porphyroblasts of garnet. Cutting the limestone are several small irregular masses of granite. These are up to 2 metres thick and often send out thin veins up to 5 cm. thick into the adjacent rock. The best

zoned skarns are seen at the junction of the most southerly of these granite bodies, and its associated veins, with the limestone in the upper part of the quarry wall, and good specimens may be collected from the rubble which partly fills the quarry. No development of skarn was seen around the large granite vein in the central part of the quarry, but a thin

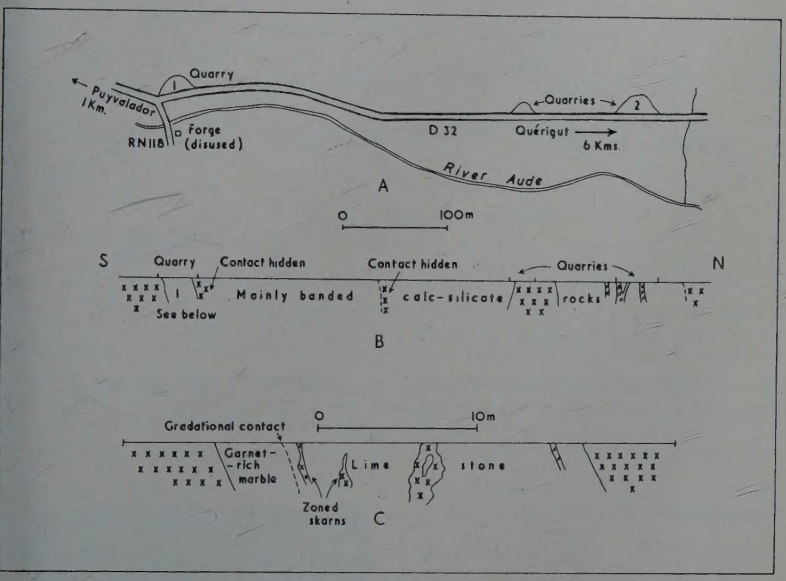

FIG. 2. Sketch-plan and sections of quarries near Puyvalador. A and B. Plan and section along road (D 32) where belt of interbanded calcareous and siliceous rocks meets the Aude valley. C. Section across large quarry (quarry 1) at road junction (RN 118–D 32).

aplite vein on the northern side shows a discontinuous layer of pinkish clinozoisite at its contact with the marble.

In the accompanying figures (3–5) are shown the arrangement and composition of zones in typical skarns collected from quarry 1 (fig. 2). Noteworthy in many of these rocks is the beautifully marked regularity of the separate zones, both in the exomorphic skarn and, to a lesser extent, in the adjacent modified granite. Next to the marble wollastonite forms an almost monomineralic zone, up to 4 cm. wide, and occurs in interlocking prisms usually aligned more or less at right angles to the contact with the marble. In section (pl. XIX, fig. 1) it is seen to carry numbers of tiny shapeless crystals of diopside, which may also form a very thin zone, observable in hand specimens as a distinct greenish line,

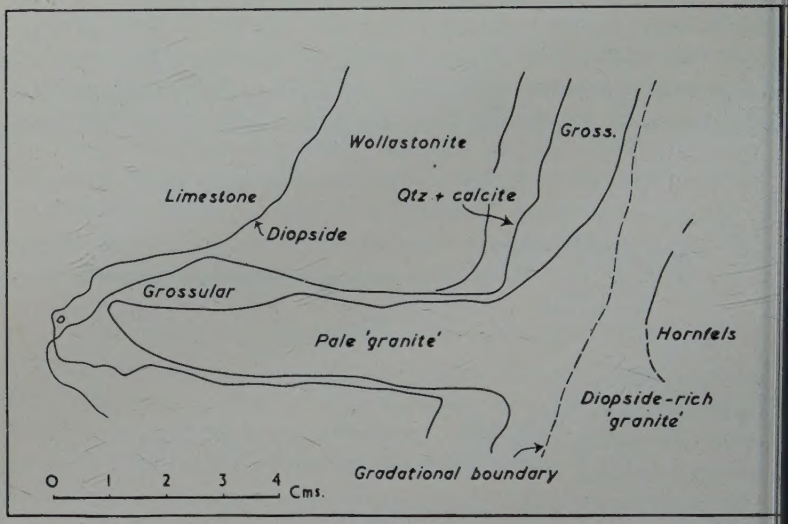

FIG. 3. Sketch of zoned skarn contact (61242), quarry at road junction north of Puyvalador, showing general arrangement of exomorphic skarn zones and modified 'granite' zones.

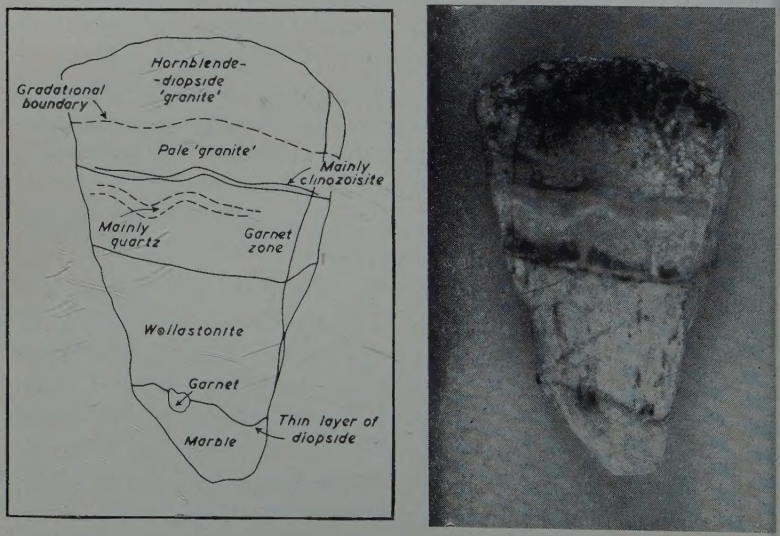

FIG. 4. Sketch and photograph of zoned contact from quarry at road junction north of Puyvalador. (Scale in centimetres.)

between the marble and the wollastonite. Other minerals in the wollastonite zone include grossular and idocrase (pl. XIX, fig. 1), crystals of which sometimes produce a brownish speckling on the surface of the zone (fig. 4).

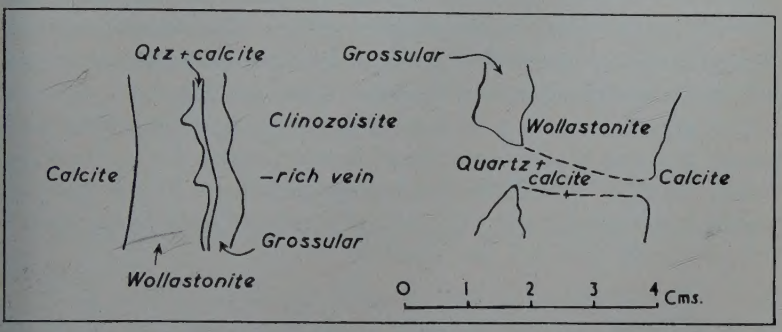

FIG. 5. Sketch showing clinozoisite-rich modified vein lined by exomorphic skarn zones, quarry at road junction north of Puyvalador.

In most specimens the wollastonite zone gives place abruptly to a pinkish-brown grossular zone, but occasionally a narrow layer in which quartz and calcite are predominant is seen between the two zones (figs. 3 and 5). Under the microscope this layer is seen to consist of a more or less equigranular mass of calcite and quartz, often unevenly distributed and usually accompanied by a little diopside. Occasionally the quartz

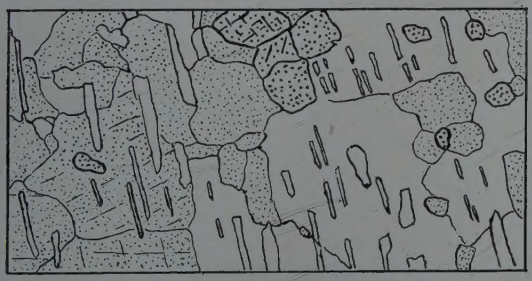

FIG. 6. Camera-lucida sketch of quartz–calcite zone between wollastonite and grossular zones (61483), showing little prismatic crystals of wollastonite within calcite and quartz. Note also small irregular grains of diopside. (Ordinary light, ×70.)

and calcite grains enclose little acicular wollastonite crystals, usually showing parallel orientation and optical continuity within the one host crystal (fig. 6).

The grossular zone is composed largely of an almost pure grossular, colourless in thin section (pl. XIX, fig. 2). It is often accompanied by a little idocrase; small, though variable, amounts of quartz, clinozoisite, diopside, and calcite may be present. Towards the outer part of the zone little irregular crystals of clinozoisite appear more constantly, and the rock grades rapidly to the modified granite. The arrangement and general nature of the modified granite zones are also indicated on the accompanying figures, and are described below (p. 718).

Here and there one or more zones may be missing, although the continuity of a zone is rarely interrupted for more than a few centimetres. Again, partial fracture of the exomorphic skarn zones by the adjacent granite is occasionally seen. Analyses of the wollastonite and grossular zones are given in table I (nos. 2 and 3).

TABLE I. Chemical data on Puyvalador contact rocks. Cols. 1–7: analyses of skarn and associated rocks. Cols. 8 and 9: figures showing gains (+) and losses (−), in grams per 100 c.c., in the marble during its replacement by the wollastonite and grossular zones.

	1.	2.	3.	4.	5.	6.	7.	8. 1 → 2.	9. 1 → 3.
SiO_2 .	4·50	49·41	49·07	73·66	57·55	70·00	61·40	+134·6	+147·7
Al_2O_3 .	1·58	2·12	16·78	13·87	15·56	15·14	16·36	+2·0	+50·4
Fe_2O_3 .	0·18	n.f.	1·58	0·32	0·81	0·74	1·91	—	+4·7
FeO .	0·55	1·42	0·75	0·28	5·26	1·90	0·73	+2·7	+0·9
MgO .	0·84	1·10	0·51	0·14	2·58	0·85	0·76	+1·0	−0·6
CaO .	53·14	44·27	29·91	6·83	11·24	2·38	12·54	−13·0	−47·0
Na_2O .	0·03	0·06	0·58	3·42	3·47	3·45	2·11	+0·1	+1·8
K_2O .	0·02	0·03	0·11	0·84	2·43	4·56	2·03	+0·1	+0·3
H_2O- .	0·02	0·02	n.p.	0·03	0·02	0·09	0·02	—	—
H_2O+ .	0·11	0·34	0·22	0·53	0·49	0·53	0·78	—	—
TiO_2 .	0·08	0·15	0·15	0·11	0·72	0·31	0·27	+0·2	+0·3
P_2O_5 .	0·06	0·06	0·02	0·01	0·19	0·11	0·15	—	—
CO_2 .	38·63	1·50	0·68	0·84	0·07	0·03	1·19	−101·7	−102·9
MnO .	0·02	0·09	0·12	tr.	0·13	0·04	0·04	—	—
	99·76	100·57	100·48	100·88	100·52	100·13	100·29	—	—
Sp. gr. .	2·72	2·97	3·26	—	—	—	—	—	—

1. Marble (61482), quarry at road junction, near Puyvalador (in this rock the figure for CaO includes 0·09 % SrO, determined spectrographically).
2. Wollastonite zone (61482), skarn in quarry at road junction, near Puyvalador.
3. Grossular zone (61482), skarn in quarry at road junction, near Puyvalador.
4. Pale modified 'granite' next to zoned skarn (61483), quarry at road junction near Puyvalador.
5. Clinopyroxene-rich modified 'granite' next to zoned skarn (61482/83), quarry at road junction, near Puyvalador.
6. Biotite-granite (61529), Puyvalador area.
7. Clinozoisite-rich vein penetrating marble (61249), quarry near Puyvalador.

(Anal. W. A. Watters.)

Of the minerals in the skarns clinozoisite shows the greatest variation in quantity. In many specimens it is present only in a few small crystals at the boundary of the grossular zone and the modified granite. Sometimes, however, it forms a thin, almost monomineralic layer on the outer side of the garnet, and grades fairly rapidly to a pale-coloured clinozoisitized 'granite' (fig. 4). In a few specimens the marble is penetrated by thin veins, which are bordered by well-developed wollastonite and grossular zones and are found to consist almost entirely of clinozoisite and quartz (fig. 5).

Mineralogy of the skarn zones.

The properties of the more important skarn-forming minerals are given below:

Wollastonite is conspicuous in pearly white prisms up to 15 mm. long. In thin section the crystals have a tabular to prismatic habit; in all the sections examined, however, no definite crystal faces were recorded. The optical properties of the wollastonite (α 1·618, β 1·632, γ 1·635, $2V_\alpha$ 35°) and the chemical composition of the zone (table I, no. 2) indicate that it lies close to pure calcium silicate. Alteration products are usually rare. In a few places, e.g.[1] 61258, it may be almost completely replaced by finely divided calcite through which the original outline and cleavage traces of the wollastonite may still be seen. In other cases, e.g. 61478, the crystals are crossed by a network of thin veins of an undetermined pale to dark brown mineral in tiny flake-like grains showing medium birefringence. This may be similar to the decomposition product noted by Osborne (1931, p. 295) on wollastonite from Marulan, New South Wales.

Grossular. In hand specimen the grossular zone is light pink through pinkish brown to brown in colour; some of this variation is due to the presence of other minerals, particularly quartz (fig. 4). The garnet itself is mainly light pinkish brown, but there are occasional small areas of considerably darker colour (fig. 4). However, the optical properties of the mineral appear to be constant, and the cause of the colour variation is not yet understood. The mineral occurs in equidimensional crystals up to 5 mm. across; these are usually shapeless except at the boundary with interstitial quartz, towards which the garnet may show well-developed dodecahedral faces (pl. XIX, fig. 2). In thin section it is

[1] Numbers refer to catalogued specimens in the Harker Collection, Department of Mineralogy and Petrology, Cambridge.

almost colourless, and the refractive index, ranging between 1·740 and 1·745, suggests a composition close to pure grossular.[1]

The Puyvalador grossular is usually isotropic, but a few of the larger crystals show anomalous birefringence (0·001–0·002) and poorly developed sector twinning (61482); occasionally mainly isotropic crystals show small irregular birefringent areas. Minute dusty inclusions are frequent. Here and there a small amount of calcite penetrates the garnet crystals along thin cracks.

Grossular is a conspicuous member of skarns from other parts of the district, as well as in many of the calc-silicate hornfelses. In general its properties are close to those recorded for the Puyvalador mineral. A few rocks carry garnet relatively rich in andradite.

Epidote minerals. A mineral of the epidote group (normally clinozoisite) is a characteristic constituent of the Puyvalador skarns. Clinozoisite is a constant, though variable, mineral in the exomorphic zones, but is probably more conspicuous in some of the modified granites and aplites. However, its properties are fairly constant in all the rocks examined. In the few relatively iron-rich skarns seen the place of clinozoisite is taken by a strongly birefringent epidote.

The clinozoisite is usually pale green; rarely, pink-tinted crystals were seen. In section it is colourless, although the crystals always have a turbid appearance owing to large numbers of tiny inclusions. Both the low birefringence and the refractive indices ($\beta = 1{\cdot}716$–$1{\cdot}726$) indicate that the mineral is low in iron. Much of the clinozoisite shows irregular zoning under crossed nicols. A calculation of the composition of the clinozoisite in an analysed rock, the clinozoisite-rich vein illustrated in fig. 5 (61249; see table I, no. 7), showed that the mineral contains about 4·8 % Fe_2O_3, corresponding to a little under 10 mol. % of the $Ca_2Fe_3(SiO_4)_3(OH)$ molecule. The refractive indices for typical grains of this mineral are: α 1·714, β 1·717, γ 1·722.

Zoisite occasionally accompanies clinozoisite in the zone immediately outside the grossular. It is always subordinate to clinozoisite in the few sections (e.g. 61251/1, 61477, 61478) where it was recorded. It occurs in long prismatic crystals showing good (010) cleavage and parallel extinction. Its refractive index ($\beta = 1{\cdot}696$) is appreciably lower than that of the associated clinozoisite.

[1] A preliminary attempt to determine the composition more accurately, by calculation from the analysis of the grossular zone (table I, no 3), indicated that the mineral is in fact close to pure grossular and contains about 5 mol. % of the andradite molecule.

Diopside is constantly present, though in small amount only, throughout the skarn zones, where it occurs usually as tiny irregular grains (normally less than 0·1 mm.) included by the other minerals (pl. XIX, fig. 1). Occasionally it may form aggregates of little crystals, generally between the wollastonite and grossular zones, but these are always local in extent. The diopside is colourless in section, and the refractive indices are: α 1·685, γ 1·716.

In a few localities a pale green hedenbergitic pyroxene (accompanying andraditic garnet) was recorded (see also Lacroix, 1900, pp. 9–11).

Prehnite is a very uncommon member of the skarn zones; where present it replaces in part the plagioclase of the modified granite next to the skarn (e.g. 61244/1, 61255/1, 61599), or fills thin veinlets penetrating the clinozoisite and grossular of the exomorphic zones.

Idocrase is a widely distributed constituent of the skarns, although it occurs in small amount only. In the wollastonite zone it is found in stout prismatic crystals, generally less than 1 mm. in length (pl. XIX, fig. 1). Within the grossular zone it forms shapeless crystals enclosed by the garnet. Sometimes a faint prismatic cleavage is visible, and there are often tiny opaque inclusions aligned parallel to the *c*-axis. Most of the crystals are a pale yellowish brown, although the colour may vary a little, even within a single grain. No pleochroism was seen. All the crystals examined are uniaxial negative, and the refractive indices vary from 1·716 to 1·722 for ϵ, and from 1·725 to 1·730 for ω, the higher values being recorded for the more distinctly brownish coloured crystals.

Most of the idocrase crystals show normal interference tints of the first order, sometimes patchily developed. A few specimens, e.g. 61245/4, show brownish-grey or greenish-brown tints, but only in very rare cases (61258) are anomalous Prussian blue colours seen. Occasionally the prisms show under crossed nicols a broad central core merging gradually towards the edge to a thin rim with slightly different interference tint. Rarely, as in 61257/5, a rough alternating concentric banding of the interference colours is seen.

Hornblende. Green hornblende forms a conspicuous member of a poorly zoned, dark-coloured skarn collected in quarry 2 from the contact between a granite vein and marble rich in garnet. It occurs in long prismatic crystals flattened parallel to (100), and is associated with abundant colourless epidote (β 1·732), calcite, pyrrhotine, and minor plagioclase and microcline.

Scapolite occurs rarely, being found only in a few specimens taken near the boundary between wollastonite and limestone (61393) and at a

contact between limestone and a small veinlet of sheared aplitic granite (61484). It is found either as large prismatic crystals or in small irregular grains, which are often intergrown with quartz. The optical properties suggest compositions in the middle part of the mizzonite range.

Sulphide minerals. As noted above, a certain amount of pyrrhotine occurs in a hornblende-rich skarn from quarry 2. It forms irregular vein-like areas, bronze-yellow in colour, cutting the other minerals and often penetrating them along thin cracks. It is accompanied by a few silvery-white grains of arsenopyrite.

Genesis of the exomorphic skarn rocks.

It has been shown above that granite veins penetrating marble in the Puyvalador area are often lined by exomorphic zoned skarns, which have evidently been formed by metasomatic replacement of the invaded rock. If it is assumed that little or no change in the volume of the replaced rock has occurred, then it is possible, knowing the compositions and specific gravities of the marble and the adjacent skarn zones, to calculate the materials added to and lost by the marble during this transformation. This has been done for one of the Puyvalador rocks (61482), and the data are given in table I (cols. 8 and 9).

It is readily seen that the main addition to the marble in the formation of the skarn has been of silica and alumina. On the other hand some lime has been lost by the marble, and this has moved out into the neighbouring granite. It is probable that the small amounts of iron oxides, alkalis, titania, and manganous oxide present in the skarn have been introduced from the granite. On the other hand, the magnesia in the wollastonite and grossular zones has probably been derived from the breakdown of subsidiary magnesian carbonate in the limestone, the presence of which is also suggested by the common occurrence of accessory diopside in the marble. However, there has apparently been some movement of magnesium within the zones, shown not only by the loss of magnesia in the grossular zone but also by the frequent development of a thin layer of diopside crystals on the inner side of the wollastonite zone (pl. XIX, fig. 1).

The figures in table I show strikingly how silica has moved into the marble in advance of the alumina, leading to the beautifully marked and regular separation of the wollastonite and grossular zones that characterizes the Puyvalador skarns. Commonly the wollastonite zone has a considerably larger volume than the grossular (fig. 3), indicating a greater addition of silica to the limestone, relative to alumina, than is

shown by the calculated figures, which refer to equal volumes of replaced marble for the two zones. However, the relative volumes of the zones may be quite variable, and the wollastonite zone itself is sometimes absent. This has been noted in particular at some other parts of the contact (e.g. in the Boutadiol valley, about 6 km. south-west of Quérigut; and near the Pic de Madrès, east of Puyvalador); the grossular zone is always present, however, and in the wollastonite-free rocks is more generally accompanied, on its outer side, by well-defined clinozoisite or clinozoisite-rich modified granite zones or by both.

The small wollastonite crystals that occur in the sporadic calcite–quartz layer (see fig. 6) indicate that some at least of the main wollastonite zone may have formed by the reaction of calcite and quartz. Experimental data on the equilibrium calcite + quartz ⇌ wollastonite + carbon dioxide have recently been given by Harker and Tuttle (1956), who have discussed also the various factors that may modify in the field the simplified conditions for wollastonite formation obtaining in the laboratory. The difficulty of determining the conditions during growth of wollastonite in nature has also been referred to by Danielsson (1950) and Pitcher (1950). Despite this difficulty, an attempt is made here to assess the possible temperature of formation of the Puyvalador wollastonite, and at the same time to suggest an explanation of the occurrence of the zone of calcite and quartz between the wollastonite and the garnet in some rocks (e.g. 61242).

If the thickness of the Devonian and Carboniferous sediments lying above the Quérigut granite at the time of skarn formation was 1750–2000 metres,[1] then the pressure due to the overlying rocks at the border of the intrusion in this locality would have been about 500 atmospheres, corresponding to a temperature of 630° C. for the production of the wollastonite (Harker and Tuttle, 1956, fig. 5). This is probably a maximum value, since, if the carbon dioxide generated during the reaction is able to diffuse away, the temperature at which wollastonite first appears (for a given rock pressure) will be lowered (Danielsson, 1950, pp. 55, 67–68). Harker and Tuttle (1956, p. 249) consider that the pressure of CO_2 may generally be comparable with the rock pressure, although it may commonly be reduced, for example by escape of the gas through fissures. In the case of the present rocks it is probable that at some stage

[1] This estimate has been taken from a small-scale section (profile 49) given in a paper by Roussel on the stratigraphy of the Pyrenees (1905, pl. 1). The thickness given above, representing that of the rocks outcropping in the district at the present day, is used with some caution in the accompanying discussion.

at least the CO_2 was able to diffuse fairly readily through the marble thus facilitating the formation of wollastonite. In some parts of the skarn, however, growth of a compact layer of wollastonite in interlocking crystals next to the marble probably hindered the escape of CO_2, the pressure of which gradually increased on the outer side of the zone, and inhibited the reaction of the calcite and silica. This gave rise to the observed calcite–quartz layer. The presence in it of tiny oriented crystals of wollastonite has already been referred to (see fig. 6), and it is probable that in parts of the zone at least the assemblage was very close to the four-phase equilibrium point for the temperature and pressure of CO_2 obtaining, and that escape of the CO_2 would have allowed the reaction to move readily to the right. However, as Harker and Tuttle point out (1956, p. 248), the effect of an additional component, in this case MgO, present in the diopside, on this equilibrium is still unknown.

There is little direct evidence regarding the order of growth of the different zones. Here and there, there are features suggesting that locally the wollastonite zone continued to grow after the formation of the major part of the garnet. For example, in 73445, a narrow tongue of fine-grained aplitic granite, continuous with the inner modified granite zone, breaks sharply through the garnet and penetrates the wollastonite zone, the thickness of which is increased around the tongue. Here the relations suggest that the narrow tongue acted as a channel through which silica continued to be introduced into the marble at a late stage in the formation of the skarn. Similar relations have been described by Pitcher for a skarn at Lough Anure, Co. Donegal, and he points out (1950, p. 138) that the supply of silica to the marble was obviously greater than could be obtained from the material of the vein itself, and that the latter must have been continuously replenished from below.

In other places, however (61483), although the pale granite has broken into both layers, sometimes penetrating as far as the marble, there is little or no thickening of the wollastonite zone, and the tongue is usually lined by a thin irregular layer of grossular. Thin sections (as well as close examination of hand specimens) show that these tongues are often highly charged with tiny grains of grossular, and the geometrical relations suggest that they have formed by intimate local penetration and replacement of the skarn zones.

The formation of grossular may be expressed by several alternative reactions, for example:

$$3CaCO_3 + Al_2O_3 + 3SiO_2 \rightleftharpoons Ca_3Al_2(SiO_4)_3 + 3CO_2,$$

or

$$3CaSiO_3 + Al_2O_3 \rightleftharpoons Ca_3Al_2(SiO_4)_3.$$

Such equations, however, may only be used in a very broad sense, as expressing the general relations between the components, and it is probable that the actual reactions were more complex, possibly involving intermediate compounds and being affected by the concentration of volatiles. Yoder (1950, pp. 232–6) attempted to synthesize grossular in dry sintering experiments on mixtures prepared in the stoichiometric proportions indicated above (as well as by possible reactions involving other constituents) but without success. He suggested, however, that grossular may form from intermediate compounds—some of which were prepared during the sintering experiments—which are presumed to be metastable.

It is probable that some at least of the grossular has formed by replacement of earlier formed wollastonite, and this is indicated by the growth of a thin zone of grossular around tongues of pale 'granite' cutting the wollastonite zone, referred to above (61483). No definite petrographic evidence of replacement of wollastonite by grossular has been found, however. One specimen (61252) shows a well-developed garnet-rich zone made up of isolated areas of grossular separated by wollastonite. Under the microscope the grossular crystals in this rock are seen to enclose occasional wollastonite prisms, but no textural evidence of active replacement of the wollastonite is observed.

On the other hand it is possible that the grossular has formed by a complex reaction between calcite, silica, and alumina. The co-existence of calcite and quartz in parts of the skarn has already been demonstrated, and the possibility remains that in places the grossular may have formed from an originally more widespread zone of calcite thoroughly impregnated with silica and alumina from the neighbouring granite. In such a case (for which there is as yet no direct evidence) growth of the two main zones would probably take place simultaneously, the formation of wollastonite and grossular being facilitated by the diffusion of the CO_2 produced by the reactions through the overlying rock.

Definite textural evidence of replacement of grossular by clinozoisite is also absent, but there can be little doubt that the formation of the sometimes abundant clinozoisite represents a gradation of the grossular zone towards to an assemblage stable in contact with the modified granite (cf. Joplin, 1935). Within the modified granite zones much of the clinozoisite and zoisite seen is due mainly to the partial breakdown of plagioclase. Nearer the marble growth of clinozoisite has been favoured not only by lower temperatures prevailing in the later stages of formation of the skarn and probable accumulation of water at the contact, but also

by the continued, though diminishing, influx of lime from the replaced marble into the marginal facies of the granite.

Description and genesis of the modified granite zones in contact with the skarn.

Against the outer side of the skarn zones at Puyvalador the granite o the veins shows a well-developed modified facies that is usually abou 10–15 cm. in width, although it may be narrower. In many specimen this marginal facies is clearly divisible into two well-marked zones, a inner, light-coloured zone (next to the skarn), and an outer, darker zon which is usually the wider of the two. However, the volume of the pal zone is increased by the little tongues of pale 'granite' that penetrat the skarn zones in many places, and the volumes of the two zones ar probably similar in many cases. Although the boundary between th zones is gradational, the transition from one to the other is usually ov a short distance (< 5 mm.), and the contrast between them is nearl always distinct in hand specimen (fig. 3).

The normal granite of the Quérigut intrusion has been described b Lacroix (1898, pp. 5–6) as made up essentially of potash felspar, olig clase-andesine, quartz, and biotite. Preliminary modal analysis several rocks from different parts of the intrusion shows that there some variation in mineralogical composition, particularly in the relati proportions of plagioclase and potash felspar, and that some of the rocks should be classed as granodiorite or quartz-monzonite rather th true granite. However, insufficient is known of this variation to permit detailed survey of the composition of the main mass.

A specimen of the granite (61529), from near the Puyvalador area, is describ below, and the analysis is given in table I, no. 6. It is a coarse-grained rock, white hand specimen, showing abundant large crystals of potash felspar, 2–3 cm. lo and little flakes of biotite. Apart from the large crystals the grain-size averag about 3 mm. Under the microscope plagioclase (making up about 30 % of t volume of the rock) is seen to occur in subidiomorphic tabular crystals oft sprinkled with alteration products, chiefly sericite and zoisite. Most of the cryst examined lie between An_{36} and An_{28}; a few zoned grains (e.g. $An_{31\rightarrow 20}$) are a present. The potash felspar, microcline microperthite (20 %), is mainly clear a unaltered and occurs in irregular plates carrying small inclusions of plagioclase a quartz. Cross-hatched twinning is constantly present, and most of the cryst show enclosed streaks of vein microperthite. Quartz (*ca.* 40 %) carries numerous t translucent inclusions and is always in irregular interstitial crystals showing str shadows. Biotite (8 %) is the only ferromagnesian mineral present; it shows marl pleochroism from α light straw yellow to γ dark reddish brown. Here and there i partially altered to pale greenish pennine. It contains numerous tiny semi-opa needles (rutile ?), which often form a criss-cross pattern in basal sections. Access

inerals are apatite, a little granular sphene, and zircon. A single crystal of mus-vite was seen. In sections of the granite from some other parts of the intrusion the otite is accompanied by a small amount of green hornblende, usually occurring irregular clots associated with plagioclase (basic oligoclase or acid andesine).

As with some other parts of the intrusion near the contact with lcareous rocks the granite veins in the Puyvalador area often carry ornblende, but there is no direct evidence to show that this has formed ecause of the incorporation of calcareous material within the granite agma. In the contact areas examined from this and other parts of the strict the characteristic change in the ferromagnesian minerals near ie actual contact is the appearance of a colourless diopsidic augite. he presence of pyroxene near the contact is exemplified by the outer id darker zone of modified granite found in many of the veins at uyvalador. The rock is medium-grained, and is made up essentially microcline, acid andesine, interstitial quartz, and a colourless or very ale green diopsidic pyroxene, which, with the actinolitic amphibole rming the cores of some of the larger crystals, makes up to 20 % of the olume of the rock.

In typical specimens (61245, 61482) the approximate mode is as follows: agioclase 30–40 %, microcline 25–30 %, quartz 5–8 %, and diopsidic pyroxene d amphibole 15–20 %. The texture shows considerable variation; sometimes the inerals are evenly distributed in small subidiomorphic crystals (0·50–0·75 mm.); other cases the pyroxene tends to occur in clusters of prisms, or, with the plagio-ase, is enclosed by large shapeless crystals of microcline (pl. XIX, fig. 3). From its xture and the relative amounts of plagioclase and potash felspar the rock is obably best termed a pyroxene-rich monzonite or quartz-monzonite.

The plagioclase lies within the sodic half of the andesine range, and on the whole probably a little more calcic than the plagioclase of the normal granite of the rusion. It occasionally shows slight irregular zoning, or it may be surrounded by very thin rim of more sodic composition. The amount and distribution of altera-n products of the plagioclase are rather variable. Most crystals show tiny inclu-ns of zoisite and sericite, but occasionally they contain relatively large shapeless ains of clinozoisite, and in one or two places the plagioclase is pseudomorphed by ne-grained mass resembling saussurite. The microcline is usually perfectly fresh; is only slightly microperthitic. Quartz is always in rather small interstitial rstals. The principal ferromagnesian mineral is an almost colourless pyroxene 1·698–1·700) forming squat subidiomorphic prisms that are often collected into egular groups up to 5 mm. across. In many cases the pyroxene is mantled around irregular core made up of interlocking crystals of a pale green to almost colourless inolitic amphibole (β 1·651) (pl. XIX, fig. 3), and there is little doubt from the ture that the pyroxene has developed later than the amphibole. There are a few ttered flakes of biotite. Both the mica and the amphibole show numerous little luded granules of sphene, which may also form large idiomorphic crystals or rims und iron ore grains. Accessory minerals are iron ore, apatite, and rarely zircon. Towards the inner part of the above zone the dark minerals decrease in amount,

and with the more widespread appearance of decomposition products of the plagi clase, and increase in quartz, the rock grades rapidly into the pale zone. This formed essentially of plagioclase (45–55 %) and quartz (35–45 %) (pl. XIX, fig. The plagioclase always carries numerous small grains of zoisite and less commo sericite. Potash felspar (< 10 %) varies somewhat in amount in different parts the zone. On the side against the skarn zones shapeless crystals of clinozois usually appear, and, as noted earlier, these may be abundant enough to form a th almost monomineralic layer against the grossular. If this zone is absent the p 'granite' sometimes carries a few small grains of garnet at the margin of the gross lar zone. On the other side small prisms of diopside begin to appear as the conta with the pyroxene-rich modified zone is approached.

The plagioclase of the pale 'granite' zone ranges between An_3 and An_{10}. T zoisite it contains is in the form of numerous tiny prisms or rod-like crystals w straight extinction, but in many places small irregular grains or clusters of gra showing properties typical of clinozoisite occur. The potash felspar forms sm interstitial crystals or occurs as a thin rim around some of the plagioclase grai The quartz is always in irregular crystals with numerous tiny semi-opaque inclusi (pl. XIX, fig. 4).

The analyses of typical specimens of these zones are given in table I, nos. 4 and

It is difficult to determine the possible amounts of the constituen given up by the granite veins to the adjacent marble in the formation the skarns, since no reliable analysis of an unaltered vein is availab and, in some cases at least, the veins differ somewhat in mineralogi composition from the adjacent granite of the intrusion. Even w analytical data it may be very difficult to compare the original unmo fied rock with the modified facies, when the latter often shows a d ferentiation into two well-marked zones.

In the veins examined there often appears to have been a pronoun migration of silica towards the boundary with the skarn. This is brou out by the petrography of the modified granite and the enrichment the pale 'granite' zone in quartz, and is strikingly shown by compar the analyses of the two zones (table I, nos. 4 and 5). That silica c tinued to migrate into the marble up to a late stage in the formation of skarn was suggested above. In one or two specimens the reaction tween the marble and the granite has possibly been modified by terchange of material between the granite zones and a fine-grai plagioclase–diopside–biotite hornfels, small amounts of which are s on the outer side of the pyroxene-rich modified zone (fig. 3). Some li and magnesia may have been gained by the modified granite from t source; however, the hornfels is sharply bounded against the pyroxe rich zone, and there is no apparent sign of interchange between the t rocks.

The migration of alumina from the modified granite to the marble

less easy to detect; the main mineralogical change reflecting the loss of alumina in the marginal granite is the crystallization of a pale diopsidic augite rather than hornblende. Migration of alumina towards the skarn contact probably also aided the crystallization of clinozoisite there in many rocks.

It was shown above that lime had been lost by the replaced marble, and the petrography and chemical composition of the modified zones show that some of this at least has been taken up by the granite. The lime shows a marked concentration in the monzonite zone (see table I, no. 5), and it is obvious that the main movement of lime within the modified granite has been opposed to that of silica. The gain in lime of the modified granite is attested by the following mineralogical changes: the appearance of pyroxene, often seen to be mantled around amphibole (pl. XIX, fig. 3); the gradual increase of clinozoisite towards the skarn zones; and the appearance of abundant accessory sphene developing at the expense of biotite, hornblende, and iron ore.

In some rocks the gradual increase in the amount of clinozoisite may be followed stage by stage to a final product formed essentially of clinozoisite and quartz. Excellent examples of such rocks are found in thin veins penetrating the marble (e.g. 61249; see fig. 5). Although these are lined by thin exomorphic skarn zones the mineralogy of the rocks (which may contain up to 55 % clinozoisite) and the chemical analysis of a typical specimen (table I, no. 7) suggest that more lime has been taken up by the vein than would be available from the marble replaced by the skarn lining the veins. It is possible, therefore, that these veins have formed channels into which some of the lime lost at other parts of the contact has been concentrated.

Other localities along the contact zone (e.g. Boutadiol valley, Pic de Madrès) often show hornblende-granite at the limestone boundary. In general the same mineralogical changes as recorded in the Puyvalador contacts are seen in these rocks. The appearance of a thin diopside-bearing granite zone between the skarn and the hornblende-granite is sometimes very conspicuous (e.g. 61376). However, marked differentiation of the marginal facies is seldom seen away from the Puyvalador locality, although clinozoisite may often form a definite layer against the grossular.

Comparison of the Puyvalador contact rocks with those of other areas.

Skarn assemblages analogous to those of Puyvalador have been described from many localities, e.g. Marulan, New South Wales (Osborne,

1931), Ben Bullen, New South Wales (Joplin, 1935), Barnavave, Ireland (Nockolds, 1937, 1950), and Ballachulish (Muir, 1953). However, these localities rarely show the often perfect skarn zonation characteristic of the Puyvalador rocks. In this respect the latter may be compared with the zoned skarns described by Tilley (1948, 1949, 1951) from the contact of Cambrian dolomites with the Beinn an Dubhaich granite in Skye. The Puyvalador skarns are comparable with some of the primary skarns of this area, but the development of ore skarns on the limestone side of the primary skarns is generally absent in the Quérigut area. Tilley (1949) has shown that at two localities in Skye (Kilchrist and Camas Malag) the grossular–wollastonite type of skarn found there has gained lime, which has migrated outwards from the dolomitic limestone; at the same time the magnesia eliminated from the skarn has moved inwards beyond the diopside zones of the skarn. In the Quérigut contacts, however, there is evidence of marked reciprocal reaction between the marble, which is non-dolomitic, and the granite, and the effects of the addition of lime to the marginal granite are particularly well shown. Analogous effects of reciprocal reaction are also seen at the other localities mentioned above.

At the Skye localities the effects of the essentially one-way transfer of material, especially alumina, from the granite to the dolomitic limestone, are expressed in the development of a marginal alkaline granite facies rich in potash felspar and containing alkali-bearing clinopyroxene and a little arfvedsonite. The marginal granite, however, is not noticeably desilicated, despite the transfer of silica to the adjacent skarn, and it is probable, as suggested by Nockolds (1950, p. 23), that such desilication only becomes apparent when the granite occurs as apophyses in the limestone. This is also suggested by different occurrences in the Quérigut district: at Puyvalador the modified granitic rocks showing marked desilication occur near the edges of veins penetrating the marble, and veins with quartz in only accessory amounts (e.g. 61388) are seen in the limestone in the Laurenti area, one km. south of the lake (fig. 1). At other parts of the contact (e.g. Boutadiol valley, Pic de Madrès), where a much larger mass of granite is against the marble, the marginal granite shows little or no desilication.

The alkaline modified granites that appear at the contacts in Skye, Barnavave, and Ballachulish show a marked enrichment in potash felspar relative to the normal granite. This has not been recorded definitely for the Puyvalador rocks. Nevertheless, in the Laurenti area veins (e.g. 61373, 61388, 61391) cutting the marble between $\frac{3}{4}$ and 1 km.

south of the lake show a high content of microcline, and although these rocks have not been studied in detail it is probable that they are analogous to the potash-rich rocks from Ballachulish, Barnavave, and Skye. These three localities resemble Puyvalador in the appearance of clinopyroxene in the modified facies. The pyroxene from the Skye and Barnavave rocks is alkali-bearing, but at Ballachulish no excess of soda over alumina in the marginal granite is found, and the pyroxene is a normal non-alkaline type. The only effect possibly attributable to the accumulation of a small quantity of soda-rich material in the final stages of consolidation of the monzonitic facies at Puyvalador is the presence of a few little wisps of a blue-green amphibole at the edges of some of the pyroxene crystals.

In the second main variety of primary contact skarns at Skye, namely the hedenbergite–plagioclase skarn, the associated granite shows evidence of incorporation of lime and is enriched in plagioclase. At Puyvalador the influx of lime into the granite has not resulted in an increase in the plagioclase content, owing apparently to the simultaneous migration of alumina to the skarn, and much of the lime gained has been taken up in the formation of pyroxene.

Summary and conclusions.

The probable sequence of events in the aureole of the Quérigut granite at Puyvalador may be briefly described as follows:

Contact metamorphism of the limestones and interbedded calcareous and siliceous rocks during the intrusion of the granite was followed by metasomatic replacement of the marble at many parts of the granite contact, with the production of skarns, up to 10 cm. wide and often zoned. From the temperature of formation of the wollastonite (above, p. 715) this has taken place under pneumatolytic conditions, and has involved the interchange of silica, alumina, and lime between the granite and the replaced marble. Finally, accumulation of volatiles in the crystallizing modified granite facies adjacent to the skarn facilitated the formation of abundant clinozoisite, the presence of which is a characteristic of the Puyvalador skarn contact rocks. Minor mineralogical changes connected with the final stages of contact activity were the production of very small amounts of prehnite in the modified granite, and the introduction of iron-rich solutions into the skarn at one or two places, leading to the formation of iron ores (magnetite and pyrrhotine) cutting the earlier minerals (see also Lacroix, 1900, pp. 9–11).

The very regular and often unbroken zonation of the skarns points strongly to a completion of movement in the greater part of the underlying granite during the period of formation of the skarn rocks and the modified granite zones. However, the presence of occasional fracturing of the skarn indicates that local minor movements in the magma continued to a late stage in the evolution of the contact rocks. It is not known to what extent this fracturing of the skarn may have further modified the composition of the adjoining granite.

In none of the contact rocks studied and described above has evidence been found of the production of hornblende-rich basic rocks, as was suggested by Lacroix (1898, 1900) for some parts of the contact in the Quérigut area. The features described show that the marginal facies of the granite in contact with the marble is developed on a small scale only, and shows the formation of pyroxene rather than hornblende.

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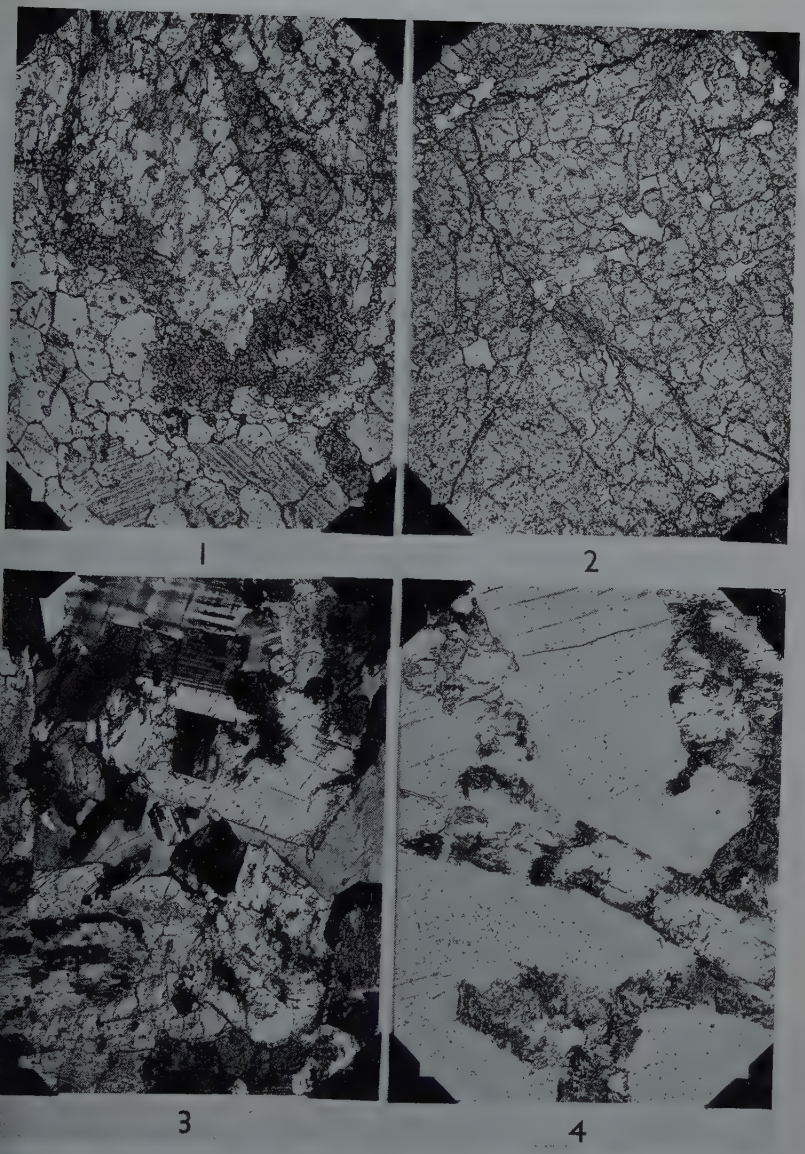

W. A. WATTERS: ZONED SKARNS FROM THE PYRENEES

Explanation of Plate XIX.

Puyvalador skarns and associated modified granite facies.

Fig. 1. Boundary between marble and wollastonite zone (61482). Note tiny cluded crystals of diopside, which also forms a very thin layer along the edge of the arble. The wollastonite zone shows a large prismatic crystal of idocrase. (Ordinary ht, × 13.)

Fig. 2. Grossular zone, showing subsidiary quartz (61482). Note occasional decahedral faces developed where the grossular is in contact with quartz. The ne often shows crystals of diopside, clinozoisite, and calcite (not seen in the otograph). (Ordinary light, × 13.)

Fig. 3. Pyroxene-rich modified 'granite' facies (61482). Note prismatic crystals plagioclase and pyroxene enclosed by microcline. The core of the large pyroxene ystal is formed of interlocking prisms of very pale green actinolitic amphibole. her minerals are quartz and biotite (not seen in the photograph) and sphene. rossed nicols, × 13.)

Fig. 4. Pale 'granite' zone (61242). Mainly quartz and acid plagioclase (albite albite–oligoclase), which nearly always contains numerous little rods and prisms zoisite. The rock may contain an appreciable, but variable, amount of potash spar. (Ordinary light, × 13.)

Zeophyllite.

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[Read 28 March 1957.]

Summary. A new analysis gives the formula $Ca_4Si_3O_{12}F_2H_6$; the specimen w from Alter Berg, Leitmeritz, Bohemia, and also contained apophyllite. The un cell is triclinic, but all reflections can be indexed on a geometrically monoclinic c which is *c*-face centred and has *a* 16·2, *b* 9·34, *c* 13·2 Å., β 113°, $Z = 6$, princip cleavage (001). This cell could also be described as having a hexagonal base of si 9·34 Å., but with *c* tilted through 23° from the vertical in the plane of one of t sides. A dehydration curve showed a step at 300–400° C., corresponding to loss two-thirds of the water. The remaining water is lost between 400° and 940° together with some fluorine, the latter probably as HF. Crystals heated at 45 600° C. retain the hexagonal-based cell, but d_{001} shrinks from 12·1 to 11·1 Å. a there is evidence of much stacking disorder of layers normal to *c*. Single cryst heated at 700° C. show ordered transformation, giving cuspidine twinned in th orientations as the only recognizable product. At 940° C. a mixture is obtain mainly β-$CaSiO_3$ with some β-Ca_2SiO_4.

ZEOPHYLLITE was discovered at Leitmeritz, Bohemia, by Pelik (1902 *a* and *b*), associated with apophyllite and zeolites in cavit in basalt. Pelikan considered the composition to be $Ca_4Si_3O_{11}H_4F_2$. T crystals were lamellar, and from etch figures he concluded that th were trigonal with cleavage (0001). Optically they were uniaxial neg tive with ω near 1·52, though towards the edges the crystals appea biaxial with 2E up to 27½° (2V up to 17°). Some goniometric obser tions were recorded by Pelikan, and also by Koechlin (1934), both whom believed that the crystals were probably rhombohedral. Lars (1921) gives data for the uniaxial form: $\omega = 1{\cdot}565$, $\epsilon = 1{\cdot}560$. X-ray studies have been reported.

The present investigation was made on crystals kindly provided the Geology Department of Aberdeen University. The specimen w labelled: Zeophyllite, Alter Berg, Leitmeritz, Bohemia; Aberdeen 61 Crushed material consisted of elongated flakes, which were stria parallel to their length (fig. 1). They were optically negative, with birefringence. The refractive index for light vibrating in the cleav

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plane was 1·568–1·569. They gave negative acute bisectrix figures with 2V varying between 0° and 10° from one crystal to another. Their extinction was indefinite, but γ was approximately parallel to the length of the flake and α was roughly normal to the cleavage. The tips of some crystals were opaque and had a lower refractive index; X-ray results

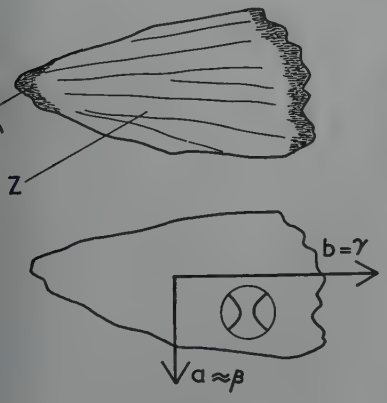


FIG. 1.

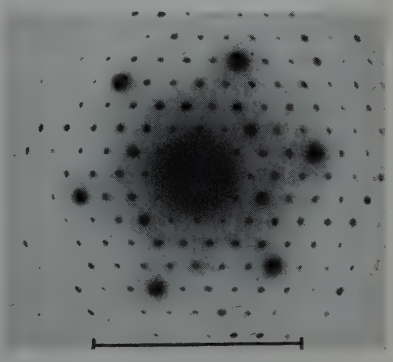


FIG. 2.

FIG. 1. *Top*: sketch of typical flake. Z = Zeophyllite. A = Apophyllite. *Bottom*: optical orientation.

FIG. 2. Electron diffraction pattern from zeophyllite crystal lying with (001) normal to the beam. Horizontal line represents 1 Å.$^{-1}$.

indicated that these ends consisted partly or wholly of apophyllite. The general appearance of our specimen agreed with Pelikan's original description except for the value of ω, which is close to that given by Larsen. The identity of the material was also confirmed by the chemical analysis (discussed later; table II) and by the correlation of the X-ray results with the earlier goniometric data.

X-ray and electron diffraction investigation. X-ray single-crystal photographs showed that the crystals were triclinic, although all reflections could be indexed on a *c*-face centred, geometrically monoclinic cell. Referred to the monoclinic axes, the photographs taken were oscillations about *b*, rotations about *b* and *c*, and *h*0*l*, *h*1*l*, *h*2*l*, *h*3*l*, *h*4*l*, and *hk*0 Weissenbergs. Electron diffraction patterns of flakes lying with their cleavages perpendicular to the beam were obtained using a Metropolitan Vickers type EM3 electron microscope.

The monoclinic cell has $a = 16{\cdot}2$, $b = 9{\cdot}34$, $c = 13{\cdot}2$ Å., $\beta = 113°$, elongation *b*, cleavage (001). β was found less accurately than the 100 and 001 spacings; d_{100} was $14{\cdot}80 \pm 0{\cdot}05$ and d_{001} was $12{\cdot}13 \pm 0{\cdot}10$ Å. The

b-oscillations showed that the crystals were only geometrically monoclinic. A triclinic cell can be chosen with $a = b = 9{\cdot}34 \pm 0{\cdot}05$, $c = 13{\cdot}2 \pm 0{\cdot}1$ Å., $\alpha = 90{\cdot}0° \pm 0{\cdot}1°$, $\beta = 110° \pm 2°$, $\gamma = 120° \pm 0{\cdot}1°$. This cell can be described as hexagonal with $a = 9{\cdot}34$ Å., and a tilted *c*-axis. The pseudo-hexagonal character is shown by the electron diffraction patterns (figs. 2 and 3) which represent a projection of the *hk*0 section of the reciprocal

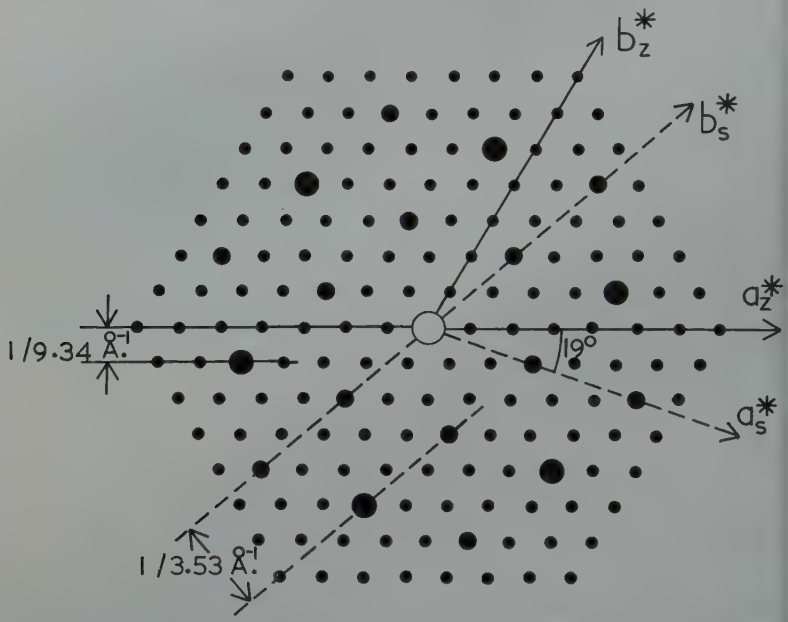


FIG. 3. Drawing from fig. 2, showing the relation of the sub-lattice formed by the strong reflections to the reciprocal lattice of zeophyllite. a_z^*, b_z^*, projections on (001) of triclinic reciprocal axes of zeophyllite. a_s^*, b_s^*, projections on (001) of reciprocal axes of the sub-lattice.

lattice on (001). The strong reflections form a sub-lattice with pseudo-hexagonal $a = 3{\cdot}53$ Å., which is similar to the *a*-axis of calcium hydroxide (3·59 Å.).

X-ray powder data were obtained (table I) using 6 cm. and 11·46 cm. diameter cameras with Cu-$K\alpha$ radiation. It was found impossible to select specimens entirely free from apophyllite. The apophyllite lines were detected by comparison with standard photographs; also, in specimens not too finely ground they were more spotty than the zeophyllite lines. Removal of the opaque portions of the flakes before crushing diminished the intensity of these lines but did not entirely eliminate

hem. Lines due wholly or partly to apophyllite are asterisked in table I. An attempt was made to index the data, but to do so was impracticable or all but the longest spacings because of the large number of reflections

TABLE I. X-ray powder data.

d, Å.	*I*	*d*, Å.	*I*	*d*, Å.	*I*
12·03	vvs	2·832	mw	1·953	vvw
7·97	vw*	2·702	w	1·913	vw
6·09	s	2·648	ms	1·858	ms
4·66	vvw*	2·547	ms	1·832	w
4·36	w*	2·431	vw	1·771	s* (d)
4·05	mw	2·344	w	1·756	w
3·96	w*	2·305	vw	1·735	vvw
3·71	ms	2·248	mw	1·704	mw
3·31	vw	2·196	mw	1·625	m
3·20	m	2·159	vw	1·601	vvw
3·07	m	2·070	m	1·554	vw
3·03	vs	2·025	m	1·535	w
2·946	s	1·986	w*	1·517	vw

* Indicates lines due wholly or partly to apophyllite.

ontributing to nearly all the lines; it was therefore not possible to refine he cell parameters by using the powder data.

Pelikan (1902*b*) observed faces inclined at 78° to (001) which he con-idered to be rhombohedral faces. Koechlin (1934) confirmed this and lso found faces inclined at 90° and at about 63° to (001). The 78°, 90°, nd 63° faces can be indexed respectively as $(20\bar{1})$, (010), and (100) on he pseudo-monoclinic axes. The calculated angles are 77°, 90°, and 67°.

Chemical analysis and cell contents. The results of a chemical analysis re given in table II, col. 2. They agree substantially with Pelikan's col. 1) but suggest a higher water content. An X-ray powder diagram f the material analysed showed slight contamination with apophyllite, ut the contamination was thought to be less than 5 %; in view of the elatively similar compositions of the two minerals, the errors caused ould not be large. The analysis was conducted on 2–25 mg. samples ithdrawn from a total sample of about 100 mg. Silica was determined y the quinoline silicomolybdate method (Miller and Chalmers, 1953*a*); lumina was determined once gravimetrically with oxine (Miller and halmers, 1953*b*) and once colorimetrically by the calcium alizarin-red ethod (Parker and Goddard, 1950). The alkali metals were determined y flame photometry and lime and magnesia by titration with EDTA.[1] he experimental error in the lime determination precluded the accurate etermination of the small amount of magnesia which was estimated to e not more than 0·5 %. Iron was tested for qualitatively and found to

[1] i.e. ethylene-diamine-tetra-acetic acid.

TABLE II. Chemical analyses and unit cell contents

	1.	2		3.	4.	5.
SiO_2	38·84	36·0	Si	9·9	9·0	9
Al_2O_3	1·73	1·5	Al	0·5	0·4	—
Fe_2O_3	0·10	nil	Fe	0·1	nil	—
MgO	0·17	<0·5	Mg	0·1	<0·2	—
CaO	44·32	46·3	Ca	12·2	12·5	12
Na_2O	0·38	0·48	Na	0·2	0·2	—
K_2O	0·24	0·09	K	0·1	<0·1	—
H_2O	8·98	11·0	H_2O	7·7	9·2	9
F	8·23	7·49	F	6·7	6·0	6
	102·99	102·9	O (not as H_2O)	29·7	28·4	27
Less O for F	3·47	3·2				
	99·52	99·7				
Sp. gr.	2·763	2·747				

1. Pelikan (1902*b*). Analyst, E. Zdarek.
2. This investigation. Analyst, R. A. Chalmers.
3. Atomic cell contents calculated from analysis 1.
4. Atomic cell contents calculated from analysis 2.
5. Theoretical atomic cell contents for $3[Ca_4Si_3O_9F_2.3H_2O]$.

be absent. Fluorine was determined by a modified Willard and Winter distillation and spectrophotometric titration with thorium nitrate solution (Hollingworth, 1957). Total water was measured as the loss in weight of a sample fused with dry sodium tungstate, which was used as a flux and as a retainer for fluorine. Tests on opal glass (F = 5·7 %) indicated that sodium tungstate did act as a fluorine-retaining flux. A Penfield determination of water on a separate sample, using bismuth oxide as retainer for fluorine, gave a slightly higher result for the water content, but the sample used was found to have contained some apophyllite, which would account for the difference.

The specific gravity was determined by suspension in bromoform–benzene mixtures. The atomic contents of the triclinic cell, calculated using the present X-ray data, are approximately $Ca_{12}Si_9O_{27}F_6.9H_2O$ (table II, cols. 3–5).

Dehydration. A weight-loss curve (fig. 4) was obtained on a 50 mg. sample using a semi-micro balance. The partial pressure of water vapour in the furnace was 6 mm. Except for the range 595–805° C. inclusive, constant weight was normally reached at each temperature. This was impracticable between 595° and 805° C. because the specimen lost weight steadily at each temperature at the rate of 50–60 μg. per day. The total loss at 1000° C. exceeded that found using a fluorine retainer, and the difference was attributed to loss of fluorine, as HF or as SiF_4. If no fluorine is lost below 400° C., the initial sharp rise in the curve

orresponds to the loss of two-thirds of the water, the remaining water eing lost, together with some of the fluorine, above this temperature. f these losses correspond to water present in the structure as H_2O and s OH respectively, the cell contents can be rewritten as

$$Ca_{12}(Si_9O_{24})(OH)_6F_6.6H_2O.$$

X-ray photographs were taken of samples withdrawn from the dehyration apparatus at 450°, 600°, 710°, 940°, and 1000° C. Powder photoraphs of the material heated to 450° or 600° C. showed diffuse patterns ith a distribution of intensity generally resembling that of the original aterial. Oscillation, rotation, and *h0l* Weissenberg photographs of the 50° C. product showed that traces of unaltered material were left, but hat the main constituent was a partly disordered phase with d_{001} reuced from 12·13 to 11·1 Å. Non-basal reflections were almost continuous reaks parallel to c^*, indicating stacking disorder of layers parallel to 001). The electron diffraction patterns were identical with those of the riginal material except for some changes in intensity, and showed that iclinic a and b were unchanged. The X-ray photographs confirmed this. he cell may therefore be treated as hexagonal with a 9·34, c 11·1 Å.

The 600° C. product was similar but still more disordered. The a-axis as still unchanged, but c had shrunk to about 10·3 Å. Traces of the 50° C. product with c 11·1 Å. remained. Certain changes in the relative tensities of the (hk) reflections were noticed both in the 450° and 600° C. roducts.

The specimen heated at 710° C. gave X-ray patterns of cuspidine ($Ca_4Si_2O_7F_2$); no other product could be detected. The change was dered, the plane of the flake becoming cuspidine (012) or (01$\bar{2}$). The spidine was twinned in three orientations with a-axes coinciding with e three hexagonal axes of the strong sub-lattice of zeophyllite (figs. and 3).

Powder specimens heated at 940° and 1000° C. gave X-ray patterns of $CaSiO_3$, with the strongest line of larnite.

Discussion. The present results confirm the earlier data for zeophyll-e, except for the crystal system and the water content; the formula is own to be $Ca_4Si_3O_8F_2(OH)_2.2H_2O$. A layer structure is indicated by e good basal cleavage, and especially by the behaviour on dehydration 450° or 600° C., which can hardly be explained except on the assumpon of distinct layers. Since the a- and b-axes remain unchanged at 0° and at 600° C., much of the structure in this plane must be preserved. follows that the atoms are most probably arranged in layers from

which the elements of water can be lost without breakdown of th layers. Possibly the initial shrinkage of d_{001} to 11·1 Å. is connected wit the loss of water molecules, and the second shrinkage to 10·3 Å. with th loss of hydroxyl groups. In both cases there is a successively close

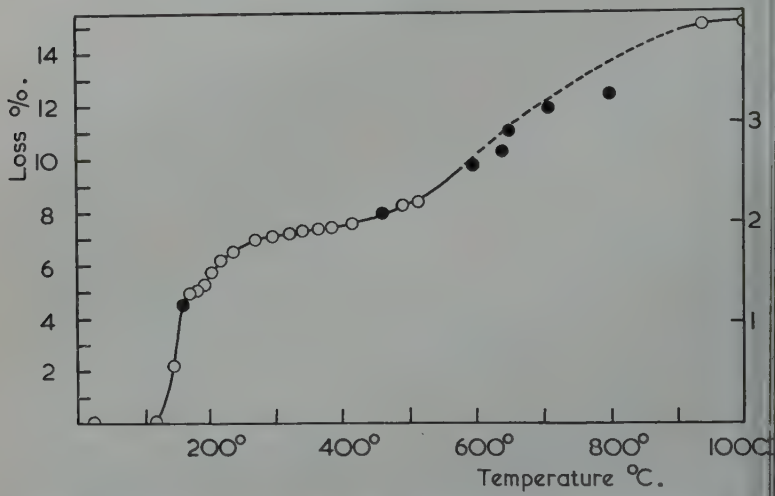


FIG. 4. Weight-loss curve for zeophyllite; partial pressure of water vapour, 6 m Open circles: equilibrium points. Solid circles: non-equilibrium points.

packing of the layers; at the same time there is loss of order in the stac ing, which leads to the non-basal reflections becoming streaks para lel to c^*.

The ratio F:Ca is the same in cuspidine as in zeophyllite. This su gests that little or no fluorine is lost below 710° C.; in agreement wi this the loss in weight at this temperature is approximately equal to t total water content. The stoichiometry of the dehydration process c therefore be written approximately as: $Ca_4Si_3O_8F_2(OH)_2.2H_2O \xrightarrow{400°}$

$$Ca_4Si_3O_8F_2(OH)_2 + 2H_2O\uparrow \xrightarrow{700°\,C.} Ca_4Si_2O_7F_2 + SiO_2 + H_2O\uparrow$$

This scheme is somewhat similar to that proposed by Brindley (195 for the dehydration of chlorites; in this case also the final produ (olivine) was twinned in three orientations at 120° apart in the plane the flake. In zeophyllite the water cannot be present in a hydroge bonded layer similar to that in montmorillonite, for the total shrinka

is too small; the water, although fairly loosely bound, presumably fits into hollows in the main structural sheets.

The reaction at 700–1000° C. could be explained as:

$$Ca_4Si_2O_7F_2 + SiO_2 \longrightarrow \beta\text{-}CaSiO_3 + 1\tfrac{1}{2}\beta\text{-}Ca_2SiO_4 + \tfrac{1}{2}SiF_4\uparrow,$$

but this gives an expected total weight-loss of 22·1 %, against the observed 15·0 %. An alternative explanation is that water is re-absorbed from the air: $Ca_4Si_2O_7F_2 + SiO_2 + H_2O \longrightarrow 2\beta\text{-}CaSiO_3 + \beta\text{-}Ca_2SiO_4 + 2HF\uparrow$. This is perhaps more likely, as it gives an expected weight-loss of 15·8 %, and a higher ratio of β-$CaSiO_3$ to β-Ca_2SiO_4 in the product, which agrees better with the X-ray result.

It seems likely that the hydroxyl present in zeophyllite is attached to calcium rather than to silicon, since it is not lost until about 600° C. (hydroxyl attached to silicon is usually lost below this temperature). If this is assumed to be so, the mineral contains silicon-oxygen sheets of empirical formula Si_3O_8. The triclinic cell contains three formula units, and consideration of its basal area suggests that there is a single Si_9O_{24} sheet in the height of the cell.

These considerations, together with the electron diffraction data, and the observed relation of orientations in the transformation to cuspidine, give some clues to the structure of zeophyllite. The sub-lattice of strong (*hk*0) reflections (fig. 3) suggests that the heavy calcium ions may occur in a pattern similar in (001) projection to that of calcium hydroxide, and from the reasons outlined above it seems likely that the calcium ions may be present in sheets. To account for the pattern completely the orientation of such sheets relative to the Si–O sheets must be the same as that of the sub-lattice to the true lattice. The basal area of the zeophyllite cell is about seven times that of the calcium hydroxide cell, which contains a single calcium ion. The unit cell of zeophyllite with 12 calcium ions therefore probably contains two such sheets, one or both of which must in some way be defective in calcium.

The same conclusions may be reached by considering the transformation to cuspidine, the structure of which has been determined by Smirnova, Rumanova, and Belov (1955). The unit cell is monoclinic with *a* 7·53, *b* 10·41, *c* 10·83 kX, β 69° 56′. The Si_2O_7 groups have their long axes parallel to *a*, and there are bands of $Ca(O,F)_6$ octahedra parallel to (012) and (01$\bar{2}$) (fig. 5). These bands are equivalent to strips of calcium hydroxide sheets, without the hydrogen atoms and with partial replacement of oxygen by fluorine. The occurrence of three orientations in the cuspidine formed from zeophyllite confirms that the sheets in zeophyllite must have hexagonal or trigonal symmetry. Half

of the Ca-containing bands in the cuspidine could be formed directly from the calcium sheets in the zeophyllite; both the plane of the sheets and the orientation within the plane correspond. The observed relation of orientations in the transformation further suggests that the Si_3O_8 sheets in zeophyllite may be capable of giving Si_2O_7 groups with their long axes parallel to each of the a-directions in the sub-lattice.

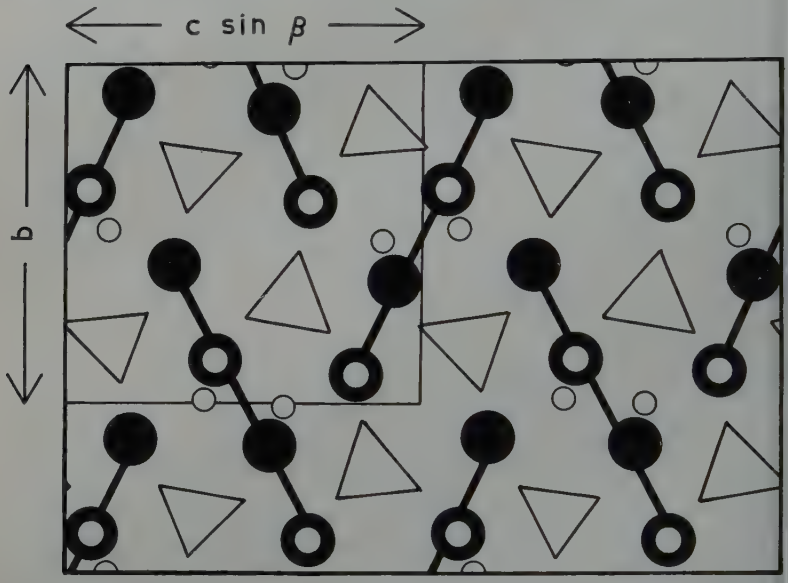
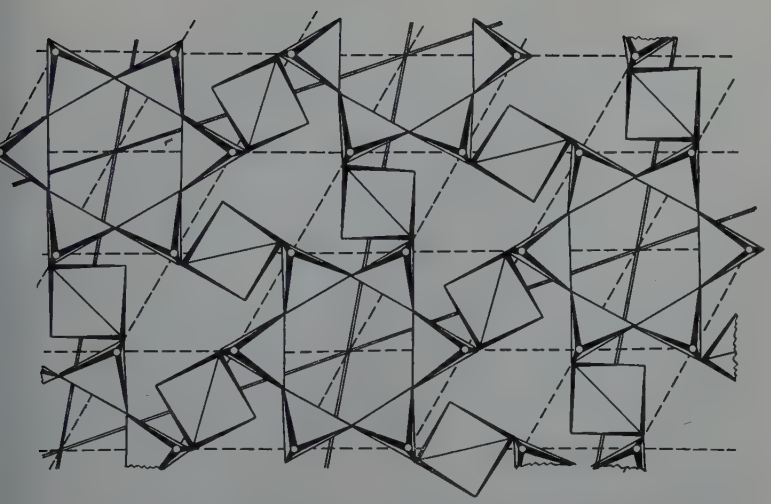


FIG. 5. Structure of cuspidine (after Smirnova, Rumanova, and Belov, 1955) projected on (100). Large full circles represent Ca ions at heights 0 and $\frac{1}{2}$; large open circles, Ca ions at heights $\frac{1}{4}$ and $\frac{3}{4}$; small circles, fluorine; triangles, Si_2O_7 groups viewed end-on; heights not indicated.

The empirical formula of the Si_3O_8 sheets could most obviously be attained by a fusion of rings of six and nine tetrahedra. Experiments with scale models suggested that the sheets might be of the form shown in fig. 6. The tetrahedra forming the six-membered rings are all at the same level, and are joined to each other by bridge tetrahedra at a different level. Points in favour of such a sheet include: the hexagonal base of the unit cell is of the right size, and bears the correct relationship to the postulated Ca–O sheets; portions of the Si–O sheets resemble the types of chain characteristic of calcium silicate minerals such as wollastonite, xonotlite, and tobermorite, with a repeat unit of three tetrahedra in a distance of 7·3 Å.; and Si_2O_7 groups in the correct orientation.



IG. 6. Suggested arrangement of SiO_4 tetrahedra to give a sheet of empirical formula Si_3O_8. Heavy lines outline lattice of zeophyllite (a = 9·34 Å.); broken lines outline sub-lattice (a = 3·53 Å.).

or the formation of cuspidine can be selected from the six-membered ings.

It is hoped to test these hypotheses by a structural analysis.

Acknowledgement. We thank Mr. R. P. Hollingworth (Univ. of Durham) for the uorine analysis.

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The system diopside–anorthite–åkermanite.

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Summary. The high-temperature phase equilibrium relationships in the system diopside–anorthite–åkermanite have been investigated by the quenching method. This system may be regarded as essentially a simple eutectic system. The eutectic is located at the composition 9 diopside:44 anorthite:47 åkermanite (by weight) and at a temperature of 1226° C. Åkermanite is a stable phase at least as low as this temperature. Triclinic anorthite is the only form of $CaAl_2Si_2O_8$ encountered in the liquidus range of temperatures.

ONE of the most important four-component systems for the consideration of petrological and technological problems is the system lime–magnesia–alumina–silica. Much work has already been done on various portions of this system. Considerable work remains to be done before the relations in the system are fully understood.

The present investigation concerns the system diopside($CaMgSi_2O_6$)–anorthite($CaAl_2Si_2O_8$)–åkermanite($Ca_2MgSi_2O_7$), which constitutes a plane in the lime–magnesia–alumina–silica tetrahedron. Diopside, anorthite, and åkermanite are end-members in the important mineral solid-solution series pyroxene, plagioclase, and melilite, respectively. A number of investigators, including McCaffery, Oesterle, and Schapiro (1927), Janecke (1933), Nurse and Midgley (1951), Osborn, de Vries, Gee, and Kraner (1954), and Prince (1954), have indicated that pyroxene, anorthite, and melilite form a stable equilibrium phase-assemblage. But no systematic study of the liquidus relations for the diopside–anorthite–åkermanite system has thus far been carried out.

The system diopside–anorthite has been studied by Bowen (1916) and by Osborn (1942). The diagrams resulting from these two studies are in essential agreement. Osborn, however, proposes a small departure from the true binary behaviour depicted by Bowen, because of the slightly aluminous character of the diopside. The diopside and anorthite liquidus curves meet at a composition of 42 anorthite:58 diopside, and a temperature of 1274° C.

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The diagram for the system diopside–åkermanite, as presented by erguson and Merwin (1919), is that of a simple binary system. The utectic lies at the composition 42 åkermanite:58 diopside. The eutectic mperature was not specified, but appears from their diagram to be out 1365° C.

The anorthite–åkermanite system has recently been investigated by e Wys and Foster (1956). The liquidus relationships suggested a simple nary system, with the eutectic located at the composition 46 anorth-e:54 åkermanite, and at a temperature of 1234° C. No evidence in sup-ort of the claim that åkermanite becomes unstable below 1325° C. (Osborn nd Schairer, 1941) was encountered. It was also concluded that the ndency of åkermanite to abstract alumina from anorthite–åkermanite mpositions to yield a melilite solid solution is negligible.

The foregoing investigations furnished the necessary liquidus data r the three limiting sides of the system diopside–anorthite–åkermanite. hey also provided index of refraction measurements on the homogeneous asses of each of the limiting systems. In addition, they yielded reliable alues for the melting-points of the three compounds involved in the resent study. These melting temperatures are: diopside, 1391·5° C.; northite, 1553° C.; and åkermanite, 1454° C.

Methods.

Thirty compositions were selected for the determination of the quidus relations of the system under investigation. In each case a mogeneous glass was prepared from high-purity $CaCO_3$, MgO, Al_2O_3, nd SiO_2. Each mixture was rendered uniform by three fusions, each of hich was followed by quenching in water, and by fine grinding of the sulting glass. The mixtures were fused in graphite crucibles in an ax-Northrup induction furnace. Previous tests on a number of mix-res with known liquidus temperatures had demonstrated that this ethod led to no detectable departures from the calculated compositions. portion of each of the thrice-fused glasses was heated for 1–48 hours in platinum crucible in the temperature range 900°–1200° C., in order to ect substantial devitrification.

The high-temperature equilibrium relations were determined by the enching method outlined by Shepherd, Rankin, and Wright (1909). his method involves wrapping about 10 mg. of devitrified glass powder platinum foil, and holding it at a selected temperature in a platinum-ound furnace for a time sufficient to ensure equilibrium. By quenching

the charge in mercury, the phases representing the equilibrium conditio at the selected high temperature are retained at room temperature. Th polarizing microscope, aided in selected instances by X-ray diffractio analysis, permits the ready identification of these phases.

Since the silicate liquids formed from these mixtures were moderatel fluid, prolonged runs were not required to establish equilibrium in th charges. The individual runs varied, in length of time, from 30 to 5 minutes. Liquidus temperatures were established by bracketing be tween two temperatures 4° C. apart, one above and one below the actu liquidus. At suitable intervals the platinum: platinum–rhodium (10 % Rh) thermocouple was calibrated against the following reference melting points: gold, 1063° C.; diopside, 1391·5° C.; and pseudowollastonit 1544° C.

The optical constants of the three crystalline phases, diopside, a orthite, and åkermanite, as well as of their homogeneous glasses, a accurately known from previous investigations. It will suffice here t present only those characteristics which served to distinguish the phases from each other in the quenched charges.

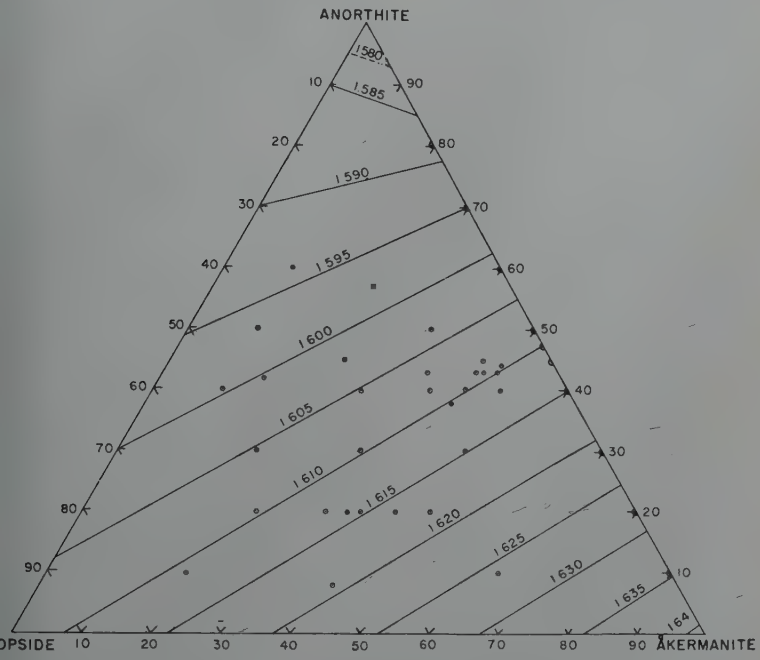
Glasses. The index of refraction chart for glasses in the system dio side–anorthite–åkermanite is presented in fig. 1. A tentative chart this type was immediately available by joining appropriate index valu of the three limiting systems previously investigated. As each of the glasses was prepared, its composition and homogeneity could be check by reference to this chart. At the same time, the essential validity of t tentative chart was confirmed.

Diopside was readily distinguished from anorthite and åkermani by its decidedly higher indices of refraction. Moderately high bir fringence and a tendency to form lath-like elongated crystals, exhibiti large (40° maximum) extinction angles, were also characteristic. Pos tive relief with respect to the glassy matrix was shown in every instanc

Crystals of *anorthite* occurred usually as fine needles which invariab possessed lower indices of refraction than the enclosing glass. Incline extinction as high as 40° was observed on these needles; interferen colours did not exceed first order grey or white. In every instance t primary crystals appeared to be triclinic anorthite; the hexagonal a orthorhombic forms described by Davis and Tuttle (1952) and by Gol smith and Ehlers (1952) were not encountered.

Åkermanite formed more or less equidimensional crystals with a pr nounced tendency towards a blocky outline. Such blocky crystals d played parallel extinction. The maximum interference colours observ

vere grey-white. Like diopside, åkermanite showed positive relief gainst the surrounding glass.



IG. 1. Isofract diagram for glasses of the system diopside–anorthite–åkermanite (weight %).

Results.

Quenching data for the thirty selected compositions involved in the resent study are presented in table I. For convenience, the data are ranged according to the identity of the primary phase obtained in the uenched charges. Only those runs which furnished limiting values of he liquidus temperatures have been included.

The data presented in table I were used to construct the phase diagram own as fig. 2. Each of the compounds diopside, anorthite, and åker- anite is seen to possess a large primary phase area. It was concluded y de Wys and Foster (1956) that åkermanite had little if any tendency abstract alumina from anorthitic mixtures to yield a gehlenite-bear- g melilite. Such an effect, had it occurred, would cause a departure om ternary behaviour in the present system. According to the work of sborn (1942), the diopside encountered in the diopside–anorthite

TABLE I. Thermal data for the system diopside–anorthite–åkermanite.

Weight % Di	An	Åk	Time (min.)	Temp. °C.	Final condition*
Primary phase diopside					
50	40	10	30	1274	Gl
			30	1270	Di, gl
43	42	15	30	1264	Gl
			30	1260	Di, gl
70	10	20	30	1350	Gl
			30	1346	Di, gl
50	30	20	30	1292	Gl
			30	1288	Di, gl
55	20	25	45	1318	Gl
			40	1314	Di, gl
30	40	30	30	1260	Gl
			30	1256	Di, gl
45	20	35	40	1308	Gl.
			30	1304	Di, gl
35	30	35	30	1278	Gl
			30	1274	Di, gl
42	20	38	30	1304	Gl
			40	1300	Di, gl
19	43	38	30	1246	Gl
			30	1242	Di, gl
30	38	32	30	1264	Gl
			30	1262	Di, gl
40	20	40	30	1308	Gl
			40	1302	Di, gl
20	40	40	30	1248	Gl
			40	1244	Di, gl
50	8	42	30	1340	Gl
			30	1336	Di, gl
18	38	44	40	1224	Gl
			30	1240	Di, gl
12	43	45	30	1234	Gl
			30	1230	Di, gl
11	43	46	50	1230	Gl
			30	1226	Di, gl

Weight % Di	An	Åk	Time (min.)	Temp. °C.	Final condition*
Primary phase anorthite					
40	50	10	30	1312	Gl
			30	1308	An, gl
30	60	10	30	1382	Gl
			30	1378	An, gl
20	57	23	30	1342	Gl
			30	1338	An, gl
30	45	25	30	1266	Gl
			30	1262	An, gl
15	50	35	30	1284	Gl
			30	1280	An, gl
10	45	45	30	1240	Gl
			30	1236	An, gl
Primary phase åkermanite					
35	20	45	30	1308	Gl
			30	1304	Åk, gl
15	40	45	45	1240	Gl
			30	1236	Åk, gl
9	43	48	30	1230	Gl
			30	1226	Åk, gl
30	20	50	40	1318	Gl
			50	1314	Åk, gl
20	30	50	30	1288	Gl
			30	1284	Åk, gl
10	40	50	30	1254	Gl
			30	1250	Åk, gl
25	10	65	30	1380	Gl
			30	1376	Åk, gl
Eutectic horizontal					
11	43	46	40	1228	Di, gl
			30	1224	Di, An, Åk
10	45	45	30	1228	An, gl
			30	1224	Di, An, Åk
8	44	48	30	1228	Gl
			30	1224	Di, An, Åk

*Di, diopside; An, anorthite; Åk, åkermanite; Gl, glass.

system was slightly aluminous. The deviation from binary behaviou resulting therefrom would carry over unavoidably into the syste diopside–anorthite–åkermanite. Since such a deviation would be onl slight the present system may for all practical purposes be regarded as simple ternary eutectic system.

Because of the simplicity of the system, the liquidus data on th thirty compositions studied were sufficient to permit the approximat location of the isotherms and boundary curves, as shown in fig. 2

The boundary curves come together at the approximate composition 9 diopside:44 anorthite:47 åkermanite, and at a temperature which has been located at 1226° C. As suggested above, this intersection point may be regarded as essentially a ternary eutectic. It is significant that no

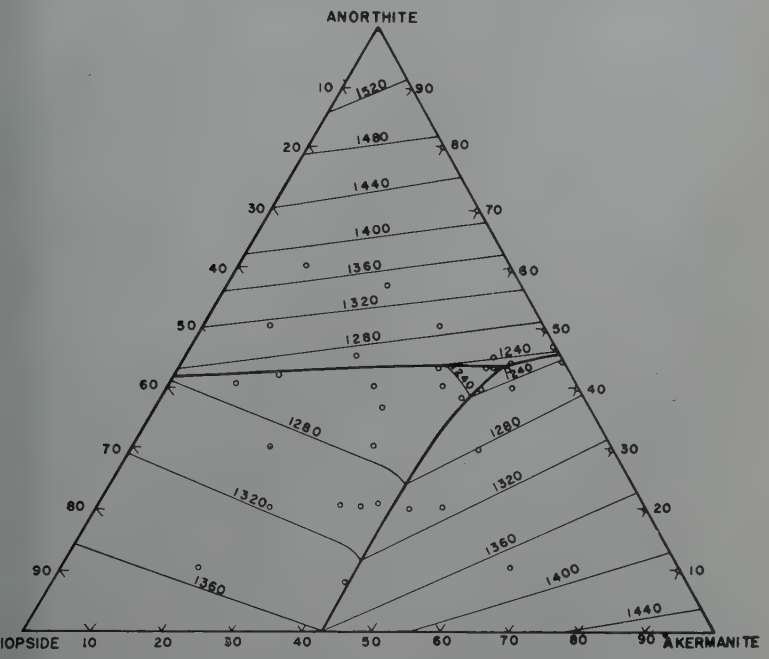


FIG. 2. Equilibrium diagram for the system diopside–anorthite–åkermanite, showing isotherms (weight %).

additional primary fields, which might reasonably be expected on the basis of the alleged instability of åkermanite below 1325° C., are found to occur in this system.

Discussion.

The experimental data of this study indicate that diopside, anorthite, and åkermanite form essentially a simple ternary system in the liquidus range of temperatures. Thus, a new plane has been added to those already known to participate in the subdivision of the system lime–magnesia–alumina–silica into subsystems. These data cannot fully explain the relationships between the pyroxenes, the plagioclase felspars, and the melilites. However, a knowledge of this system serves to fill one of the existing gaps in our understanding of them.

The system sheds some light on the problem of the stability of åkermanite. It has long been held that åkermanite becomes unstable below 1325° C. (Osborn and Schairer, 1941). This claim is at variance with earlier work (Taylor and Williams, 1935) which recorded the solid-state synthesis of åkermanite in the temperature range 800–1200° C. Recently Nurse and Midgley (1953) and de Wys and Foster (1956) have reported studies purporting to show that åkermanite does not become unstable on cooling below 1325° C. The present study lends added support to these latter claims: åkermanite was found to crystallize from appropriate liquids of the system diopside–anorthite–åkermanite at least 100° C. below the alleged stability limit; if åkermanite were unstable in the liquidus range of temperatures of this system, the primary field of some phase or phases other than diopside, anorthite, and åkermanite might have been expected; no such additional phase was encountered.

The present study was incapable of disclosing the equilibrium relationships existing well below the eutectic. It is of interest in this connexion that subliquidus studies by Harker and Tuttle (1956) have shown that below about 700° C. åkermanite breaks down into wollastonite and monticellite. At the same time their results confirm rather than contradict those of this investigation. Thus, although Harker and Tuttle indicate that åkermanite possesses a low-temperature stability limit their results confirm the stability of this phase above 700° C. Belief in its inherent instability on passing below 1325° C. must therefore be discarded.

The results of Harker and Tuttle are of further interest in connexion with the present study. They indicate that the system diopside–anorthite–åkermanite may exist as a dividing plane in the lime–magnesia–alumina–silica tetrahedron only above 700° C. Indeed, they suggest that all phase diagrams depicting the coexistence of åkermanite with other crystalline phases are valid only above 700° C., unless the intervention of solid solution phenomena lowers this limit. It would therefore appear that such additional diagrams as that of the anorthite–åkermanite system (de Wys and Foster, 1956) and the central portion of the lime–magnesia–silica system (Ferguson and Merwin, 1919; Ricker and Osborn, 1954) are valid only above 700° C.

No further data on the interrelationships between the hexagonal, orthorhombic, and triclinic forms of $CaAl_2Si_2O_8$ were obtained. Only the triclinic form, anorthite, was found to crystallize from the liquids of this system.

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The powder patterns and lattice parameters of plagioclase felspars. II.

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Summary. The angular separations (2θ (131)$+2\theta$ (220)-4θ ($1\bar{3}1$)) and (2θ ($1\bar{1}1$)-2θ ($\bar{2}01$)) for 111 analysed plagioclase felspars have been determined from diffractometer records. From An_0 to An_{70} the petrological environment has a strong influence on these separations, and if the An-content is independently known an accurate estimate of the structural state can be made from the first of these angular separations. From An_{70} to An_{100} there are only small variations in both separations with An-content and structural state. From An_{85} to An_{100} inconsistencies in the data suggest that other factors can influence any interpretation so seriously that only qualitative or semi-quantitative deductions may be made. Consideration of the angular separations and subsidiary reflections of sodic plagioclases from a wide range of geological environments has led to the novel hypothesis that in this composition region even plutonic specimens are in a metastable state with slightly disordered structures.

The value to the petrographer of the available methods for the estimation of An-content and structural state is critically discussed. Although it is thought that the standard optical curves in present use are in need of some revision, the determination of optical orientation by universal-stage methods is probably adequate for routine studies. For more detailed investigations, this may be supplemented by X-ray powder data and refractive index measurements, whilst a complete description of the felspar requires the addition both of chemical analysis and single-crystal X-ray work. In particular, the efficacy of the various X-ray methods that have been proposed is discussed, and recommendations of the most suitable method for various composition ranges are made.

In paper I (Smith, 1956) the variation of lattice parameters of sodic plagioclases with chemical composition and thermal history was presented, and on the basis of these changes simple X-ray powder methods for the estimation of structural state[2] and chemical composition were given. The variation of the cell dimensions with the An-content of the natural plagioclases from pegmatites and granites is continuous. Upon heating just under the solidus, the cell dimensions changed towards the

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[2] This term is now preferred to 'thermal state', which was used in earlier papers on the plagioclase felspars.

values given by the synthetic plagioclases, most of the specimens giving final values nearly identical with those for the synthetic specimens of the same composition. These and other results provide justification for defining high and low series of soda-rich plagioclases. Natural specimens from volcanic rocks exist in structural states intermediate between these two series. In sodic plagioclases, the effects on the lattice parameters of drastic heating and of increasing lime-content are so similar that, in practice, one of the two variables, An-content and structural state, must be known before the other can be estimated by X-ray powder methods.

This paper records the results obtained by an extension of the X-ray powder studies through the remaining composition range. The angular displacements between certain adjacent reflections have been correlated with both the An-content and the geological and thermal history of natural and synthetic specimens, but further clarification of the phase relations is thought to be advisable before the determination of the cell dimensions is begun.

It is necessary in assessing the results of the powder work to take into account the structural variations in the plagioclase series revealed by single-crystal X-ray studies (Cole, Sörum, and Taylor, 1951; Gay, 1953, 1954, 1956; Gay and Taylor, 1953; Gay and Smith, 1955; Gay and Bown, 1956; Laves, 1954; Laves and Goldsmith, 1951, 1954; Sörum, 1951), and a brief résumé of the results of this work would be appropriate at this point. Most natural plagioclases from low-temperature environments (plutonic, metamorphic, pegmatitic, &c.) give complex single-crystal patterns. The low-albite structure (with $c \approx 7$ Å. for a face-centred lattice) tolerates only a little lime-felspar in solid solution, for specimens from An_3 to An_{17} were found to consist of a submicroscopic parallel intergrowth, a peristeritic texture of low-albite and a lime-rich phase (An_{20-25}). More basic specimens gave patterns with extra weak reflections in addition to the main reflections of the albite pattern; the variations in these weak reflections distinguish one structure type from another. For lime-rich specimens from An_{70-75} to An_{80-85} (the limits are imperfectly known as yet) these additional weak reflections show that if the conventional orientation of the felspar cell is retained it has a doubled c-axis and is effectively body-centred. With increasing lime content, this structure is gradually replaced by the transitional anorthite structure (An_{80-85}—An_{90-95}) and finally by the primitive anorthite arrangement (An_{90-95}—An_{100}), both of which have a doubled c-axis. Over the range An_{20-25} to An_{70-75} the single-crystal patterns reveal pairs of weak reflections symmetrically disposed about the positions that would be

occupied by the extra reflections of body-centred anorthite; the patterns are remarkable, for the separations of these pairs are variable and apparently a linear function of composition. This intermediate structure is thought to be caused by a special kind of unmixing into lime-rich and soda-rich regions, which have been tentatively identified with body-centred anorthite and the calcium-rich component of unmixed albite-oligoclases. Recent unpublished results on specimens close to the boundaries of the intermediate composition range suggest further structural complexities.

In the high-temperature series, from An_0 to An_{90} there is only one structure type, high albite, which has a face-centred cell with c-axis $\approx$ 7 Å.; it differs from low albite in that the Si–Al arrangement is disordered. Quenched synthetic anorthite (An_{100}), however, shows the transitional anorthite structure with $c \approx 14$ Å. All natural specimens may be converted into their high-temperature counterparts by prolonged heating; most natural specimens from volcanic and hypabyssal rocks are already in states part way between the low and high forms. Apart from the region of peristeritic unmixing, the effects of heat treatment upon the diffraction patterns are known. For the intermediate specimens, the pairs of subsidiary spots gradually become weaker and finally diffuse without change of position; their complete disappearance results in the attainment of the high-albite structure. Anorthites with the primitive structure undergo a rapid and reversible inversion to the transitional anorthite structure if heated above a given temperature; this critical temperature is strongly dependent on the albite content. Calcic bytownites with the transitional structure invert less readily to the body-centred structure and then to the high-albite structure. For the more basic specimens, the reaction rates are so fast that only the most rapidly cooled natural specimens retain the high-temperature structure. Over the remaining composition range, the inversion occurs slowly and has so far proved irreversible in laboratory times.

The weak reflections that distinguish these structure types are rarely apparent on powder records, and in powder work it is convenient to use the face-centred cell with 7 Å. c-axis for the indexing of reflections.

In addition to paper I of this series, X-ray powder studies have been reported by Claisse (1950), Goodyear and Duffin (1954, 1955), and Smith (J. R.) and Yoder (1956). Claisse's work was based on a photographic study of only eight natural specimens. Goodyear and Duffin extended the work to cover a wider range of natural specimens and were also able to use synthetic specimens; they found that the synthetic and

natural specimens fell into two separate groups. In their later paper, they describe the production, by the heating of natural specimens, of felspars with reflection angles between the two groups. Unfortunately, the range of geological environments covered by Goodyear and Duffin's specimens was small and only one unheated specimen gave reflection angles indicating a transitional state. Smith (J. R.) and Yoder have examined specimens from a much wider range of environments, covering the whole composition range except the anorthite field. They found that different thermal histories cause large variations in the reflection angles, with the volcanic and hypabyssal specimens generally lying between the plutonic and synthetic ones. They concluded that 'composition determinations cannot be made by the use of the available curves based on the variation of reflection separations, because there is no *a priori* way of knowing how closely a given plagioclase is represented by a particular curve. However, given the composition of a plagioclase, the curves are useful for making an estimate of its degree of inversion towards some undefined low-temperature state.'

All the separations used by Smith (J. R.) and Yoder, and all but one of those used by Goodyear and Duffin, are effectively dependent on the angle γ^*. The values of these angular separations in the composition range An_{80} to An_{100} are almost independent of structural state and Goodyear and Duffin suggested that the composition could be determined from them. The other separation used by Goodyear and Duffin (between the peaks labelled *F* and *G*) was proposed as an indicator of structural state for this composition range. Unfortunately peak *F* is complex and Goodyear and Duffin state that 'while the *F* peak is reasonably resolved from neighbouring reflections for all the natural specimens, difficulties arise in its interpretation in the patterns of some of the synthetic materials'. In our opinion, peak *F* is not suitable for routine investigation. Fortunately, the angular separation between the well-resolved $(1\bar{1}1)$ and $(\bar{2}01)$ reflections has proved to give a similar variation for basic specimens and is to be preferred to the *F*–*G* separation. It is thought that the effectiveness of both these separations depends largely on the angle β^*.

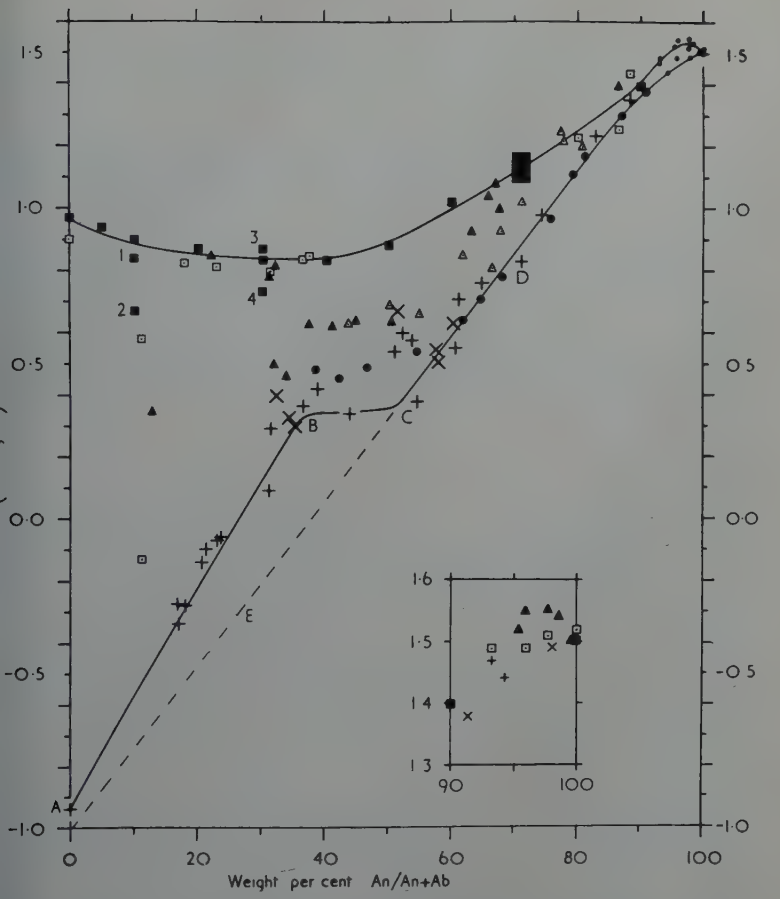
Measurements. Of the reflections that depend on γ^*, the ones most suitable for measurement in intermediate and basic specimens are 220, $1\bar{3}1$, and 131. The 220 and $1\bar{3}1$ reflections from many low albites and acid oligoclases are unsuitable because of the peristeritic unmixing and because γ^* passes through 90° with the consequent interchange in position of 220 and $2\bar{2}0$: in this composition range, the methods developed

in paper I are to be recommended. In the remaining composition range a more valuable variable is the angle $2\theta\,(131)+2\theta\,(220)-4\theta\,(1\bar{3}1)$, to be called Γ (capital gamma). As the $1\bar{3}1$ reflection lies between the other two, systematic errors in the X-ray diffractometer records are cancelled. The other separation used in the present work is the angle $2\theta\,(1\bar{1}1)-2\theta\,(20\bar{1})$, to be called B (capital beta); these two reflections are adjacent (at $2\theta_{Cu} \approx 23°$ and $\approx 22°$, respectively) and no appreciable systematic errors can occur if the diffractometer is correctly aligned. Smith (J. R.) and Yoder have measured the separate quantities $2\theta\,(1\bar{3}1)$–$2\theta\,(131)$ and $2\theta\,(220)$–$2\theta\,(1\bar{3}1)$ and Γ may be obtained by subtracting the first quantity from the second. (Unfortunately they have used the wrong sign and all their quantities should be reversed.) Many specimens used by Smith (J.R.) and Yoder have also been measured by us and an estimate of the reliability of measurement may be readily obtained. The average discrepancy in Γ, 0·015° in 2θ, is astonishingly good. Comparison of independent measurements of B indicates that an accuracy of 0·01° in 2θ has been obtained.

About half of the powder records were taken by the first author on a Norelco diffractometer at the Geophysical Laboratory; the remainder were taken on a Berthold diffractometer whose design has been severely modified by the authors and which was rebuilt by Mr. A. N. Lanham. Filtered Cu-$K\alpha$ radiation has been used for all the records. Portions of the powder pattern that contain the 131, $1\bar{3}1$, and 220 peaks have been figured by Smith (J. R.) and Yoder.

Discussion. The values of Γ for 78 natural, 16 synthetic, and 17 heated plagioclases together with the values of B for 62 natural, 9 synthetic, and 17 heated plagioclases are given in table I and plotted against An-content in figs. 1 and 2. Thirty-four values of Γ have been obtained from the data of Smith (J. R.) and Yoder, whilst 41 specimens have been measured by both sets of workers.

An attempt has been made to arrange the specimens into groups whose probable thermal histories have offered comparable opportunities for the development of an ordered structure. The groups are: synthetic; heated natural; volcanic; hypabyssal, including anorthositic specimens from the upper parts of gabbroic sills; plutonic and pegmatitic; metamorphic and charnockitic. Smith (J. R.) and Yoder separated the specimens from the stratiform layered intrusions from the other plutonic specimens and this segregation has also been retained in order to facilitate discussion. Most of the specimens can be readily assigned to one of these groups; the doubtful classification of others does not affect the validity



G. 1. The variation of $\Gamma\,(2\theta\,(131)+2\theta\,(220)-4\theta\,(1\bar{3}1))$ with wt. % An/(An + Ab). he specimens have been allocated into six groups on the basis of their origin d heat treatment, with the following symbols: ■ synthetic; □ heated natural; volcanic; △ hypabyssal; + plutonic and pegmatitic; × metamorphic and char-ckitic; ● Skaergaard, Stillwater, and Bushveld igneous complexes. Specimens belled 1, 2, 3, and 4 were synthesized hydrothermally by Smith (J. R.) and Yoder. he significance of the curves is explained in the text. Many values fall too close for dividual reproduction especially those for six specimens of compositions An_{70}–An_{72}, hich have been replaced by a rectangle elongated vertically in such a way as to proximately represent the spread of the values. Similar confusion in the region $_{90}$ to An_{100} has been avoided by representing all values by dots. An enlarged portion of this region using the proper symbols is given in the inset diagram.

the interpretations to be put forward. It will be convenient to discuss e composition ranges An_0 to An_{70} and An_{70} to An_{100} separately.

TABLE I. B and Γ values for plagioclase felspars* (for definition of B and Γ see text, p. 572).

Ref.	Specimen label	% An and Class.	B.	Γ.	Γsy.
Natural specimens					
7	KN 12	0·0—N	1·014°	(−0·94)°	
3	BM 1940, 27	11·2—N	0·988		
1	E 2 (10)	11·6—N	1·00		
10	Larsen 9	12·5—L			+0·35°
1	E 3 (73)	16·6—N	0·99		- 0·27
1	E 4 (156)	16·9—N	0·98		−0·34
7	KN 8	17·9—N	0·968		−0·28
11	SLR 596	18·9—N	0·957		
1	E 5 (94)	20·4—N	0·97		−0·14
1	E 32 (0–761–4)	21·2—N			−0·10
15	24335	21·8—L	0·898	+0·848	
7	KN 9	22·9—N	0·958		−0·07
7	KN 10	23·6—N	0·969	−0·04	−0·06
7	KN 11	31·0—N	0·935		+0·09
15	11277	31·0—L	0·892	+0·785	
1	E 28 (D—638)	31·3—N			+ 0·29
10	Larsen 8	31·8—L			+0·50
15	15472	32·1—L	0·872	+0·822	
5	S—347	32·2—P	0·903	+0·42	+0·39
10	Larsen 4	33·9—L			+0·46
5	6436	34·1—P	0·917	+0·33	+0·33
5	2270	35·1—P	0·907	+0·32	+ 0·29
1	E 6 (152)	36·4—N	0·915	+0·37	+0·36
1	E 7 (64)	37·3—L	0·875	+0·63	+0·63
20	4145	38·4—Q	0·906	+0·48	+0·48
1	E 8 (144)	38·8—N	0·880	+0·41	+0·43
10	Larsen 7	41·0—L			+0·62
20	2580	42·1—Q	0·900	+0·45	+0·45
11	EL 38–28	43·8—N	0·897	+0·34	
13	M 3174	44·1—M	0·880	+ 0·63	

Ref.	Specimen label.	% An and Class.	B.	Γ.	Γsy.
Natural specimens					
1	E 12 (97)	57·3—P	0·880°	+ 0·55°	+0·55°
1	E 13 (92)	57·7—P	0·880	+0·52	+0·51
5	2941	60·2—P	0·860	+0·66	+0·63
1	E 14 (167)	60·6—N			+0·55
1	E 16 (132)	61·1—N	0·870	+ 0·70	+0·71
17	BV 63	61·8—Q			+0·64
1	E 15 (53)	61·8—M	0·86	+ 0·87	+0·85
10	Larsen 2	63·2—L			+0·93
17	EB 41	64·3—Q			+0·71
11	SLRM 334	64·7—N	0·847	+ 0·76	
7	KN 7	65·9—L	0·800	+1·08	+1·04
1	E 17 (52)	66·3—M	0·850	+ 0·81	+0·81
6		66·9—L	0·800	+1·08	
10	Larsen 1	67·6—L			+1·00
1	E 18 (134)	67·8—M	0·850	+0·89	+0·91
17	EB 42	68·2—Q			+0·78
1	E 19 (61)	70·7—L	0·795	+1·13	+1·10
1	E 20 (22)	71·1—N	0·850	+ 0·82	+0·84
7	KN 5	71·3—M	0·805	+1·14	+1·15
1	E 22 (54)	71·3—M	0·820	+ 1·11	+1·09
1	E 21 (51)	71·7—M	0·820	+1·02	+1·03
1	E 23 (165)	74·5—N	0·825	+0·98	+0·98
17	EB 43	75·8—Q			+0·97
1	E 24 (1 B)	77·4—M			+1·24
1	E 25 (1 A)	77·8—M	0·800	+1·20	+1·23
1	E 26 (109)	79·3—Q	0·815	+1·12	+1·11
2	15	80·9—M	0·815	+1·20	
17	EB 38	81·3—Q	0·800	+1·18	+1·16
1	E 27 (166)	83·0—N	0·790	+1·24	+1·23
4	Juvinas	86·2—L	0·747	+ 1·40	

10	Larsen 5	44·6—L			+0·64°
20	1963	46·6—Q			+0·49
13	EH 20	50·3—M	0·863°	+0·69°	
10	Larsen 3	50·3—L			+0·64
1	E 9 (24)	50·9—N	0·895	+0·54	+0·54
17	C 41	51·5—P			+0·67
1	E 10 (132)	52·2—N	0·890	+0·59	+0·60
1	E 11 (151)	53·6—N	0·900	+0·58	+0·58
17	GD 29	54·3—Q			+0·54
7	KN 6	54·6—N	0·890	+0·36	+0·39
13	EH 201	55·1—M	0·863	+0·66	
Synthetic specimens					
	0	0·0—H	0·88	+0·97	
	5	5·0—D			+0·94
	10	10·0—D	0·85	+0·90	+0·90
18	S 2—1	10·0—H			+0·67
18	S 2—2	10·0—H			+0·84
	20	20·0—D			+0·87
	30	30·0—D	0·83	+0·82	+0·83
18	S 2—3	30·0—H			+0·73
Heated specimens					*Treatment.*
7	KN 12	0·0	0·915	+0·89	60°—23
3	BM 1940, 27	11·2	0·950	−0·13	60°—12
3	BM 1940, 27	11·2	0·908	+0·58	60°—24
7	KN 8	17·9	0·865	+0·82	90°—42
7	KN 9	22·9	0·895	+0·81	90°—42
7	KN 11	31·0	0·865	+0·80	90°—42
1	E 6 (152)	36·4	0·880	+0·84	90°—28
1	E 7 (64)	37·3	0·860	+0·84	90°—14
1	E 20 (22)	71·1	0·760	+1·13	300°—3

17	EB 18	86·7—Q	0·791°	+1·30°	+1·31°
14	Rhum	88·4—N	0·780	+1·35	
18	HGIF–An–53	91·3—P			+1·38
7	KN 1	93·3—N	0·757	+1·47	
12	SLR 354	94·3—N	0·730	+1·44	+1·44
16	Tarumaé	95·5—L	0·711	+1·52	
8	†	96·0—L	0·720	+1·55	
8	HK 33010901 e	97·8—L	0·710	+1·55	
19	C 50	98·1—P	0·733		+1·49
9	‡HK 350826046	98·6—L	0·707	+1·54	
2	§	99·7—L	0·700	+1·50	
Synthetic specimens					
18	S 2–4	30·0—H			+0·87
	40	40·0—D			+0·80
	50	50·0—D	0·785	+0·88	+0·86
	60	60·0—D	0·770	+1·02	+1·01
	70	70·0—D	0·740	+1·10	+1·11
	80	80·0—D	0·730	+1·24	+1·21
	90	90·0—D	0·687	+1·40	+1·40
	100	100·0—D	0·670	+1·50	+1·50
Heated specimens					*Treatment.*
7	KN 5	71·3	0·780	+1·13	100°—4
7	KN 5	71·3	0·773	n.d.	300°—4
17	EB 18	86·7	0·740	+1·36	300°—3
4	Rhum	88·4	0·713	+1·44	100°—3
7	KN 1	93·3	0·693	+1·49	180°—3
8	Hatiszyô-zima	96·0	0·713	+1·49	‖
8	HK 33010901 e	97·8	0·737	+1·51	¶
	100	100·0	0·723	+1·52	**

† Phenocrysts from olivine basalt occurring as beach sand. Hatiszyô-zima volcano, Seven-Izu Islands, Japan.
‡ Ejected block of allivalite in tuff, Hakoné, Japan. § Crystal lapilli (?), Mikyinna, Japan.
‖ ' Annealed ' 8 days at 750° C. ¶ ' Annealed ' 8 days at 850° C.
** ' Annealed ' 1 day at 1100° C.

In the first composition range, Γ varies much more widely than B and may be expected to give a more reliable indication of structural change for it should be affected proportionately less by such possible factors a solid solution of potassium and iron. Within the limits of experimenta error the plagioclases synthesized in the dry way form a continuou series. The natural specimens fall in a broad band with those fron

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* *Notes*: Values of B and Γ quoted in degrees. The column headed Γsy und natural and synthetic specimens contains data derived from the measurements Smith (J. R.) and Yoder (1956). The An-content is given as An/(An + Ab), wt. % T natural specimens have been classified into five groups on the basis of their occ rence: L, volcanic; M, hypabyssal including anorthosites from the upper levels gabbroic sills; N, plutonic and pegmatitic; P, metamorphic and charnockitic; Skaergaard, Stillwater, and Bushveld specimens.

Under the synthetic specimens, H (in the class column) indicates a hydrotherm synthesis, D a dry synthesis. The details of hydrothermal treatments may be fou in the appropriate reference, except for specimen O which was crystallized at 800° under 1000 Kg./cm.^{2} pressure for 5 days. The dry synthetic specimens were all p pared by Dr. J. F. Schairer (Geophysical Laboratory, Washington).

The column headed 'Treatment', under heated specimens, gives the temperat in degrees above 1000° C. and the time of heating in days, e.g. 300°—3 indica heating at 1300° C. for 3 days, and 90°—42 indicates 1090° C. for 42 days.

utonic rocks usually the farthest displaced from the dry synthetic ries. The volcanic and hypabyssal specimens fall into intermediate sitions, whilst there is some tendency for the values for hypabyssal ecimens to plot between the plutonic and volcanic ones. It is not sur- ising that the spread of values for each group is quite large, for each oup must embrace a wide range of conditions. Some of these speci- ens have been examined by single-crystal methods and the displace- ents of the values of Γ are in accordance with the estimates of struc- ral disorder inferred from the single-crystal photographs. Thus it ems reasonable to correlate Γ with the state of order.

For igneous specimens, the nature of occurrence is clearly of prime portance in controlling the degree of order attained by a particular spar. It is natural to assign a leading role to the rate of cooling in ew of the known effect of heating on the state of order, but other ctors such as pressure and the volatile content of the magma un- ubtedly have important effects although their relative importance nnot as yet be assessed.

When plagioclases are strongly heated, any structural order is stroyed and the completely disordered state may be approached in an parently continuous manner. After the severest heat treatments, the lues of Γ and more especially B do not quite reach those recorded corresponding dry synthetic plagioclases, and it seems likely that the ferences are significant. The reason for this is unknown, although the own solid solution of potash-felspar in natural plagioclases provides possible explanation. The broken-line curve in fig. 2 probably re- esents the limit towards which B for natural plagioclases tends as they heated until no further change occurs. A further complexity has en introduced by the work of MacKenzie (1952) who found slight acing differences in the powder patterns of albite synthesized in rious ways. Smith (J. R.) and Yoder have also reported variations for nthetic plagioclases crystallized hydrothermally at high pressure and derate temperature (points 1, 2, 3, and 4 in fig. 1). Again the signifi- nce of these observations is not clear, though the latter may represent ne slight order introduced into the structure by the conditions of rstallization.

Apart from these small variations, the powder and single crystal X-ray dies and the optical work provide justification for the definition of o limiting series of plagioclase felspars. Ideally the series are defined terms of the structural arrangement, the 'high' series corresponding to most disordered structure, the 'low' series to the structures with the

highest degree of order. In practice some more easily determinable func tion must be used. It is suggested that over the composition range An to An_{70} Γ is most suitable, for it is easily determined and provides a mor sensitive criterion for the detection of deviations from the limiting serie than any other so far suggested by X-ray and optical work. Other func tions are more suitable for the ranges An_0 to An_{20} (one or more of th

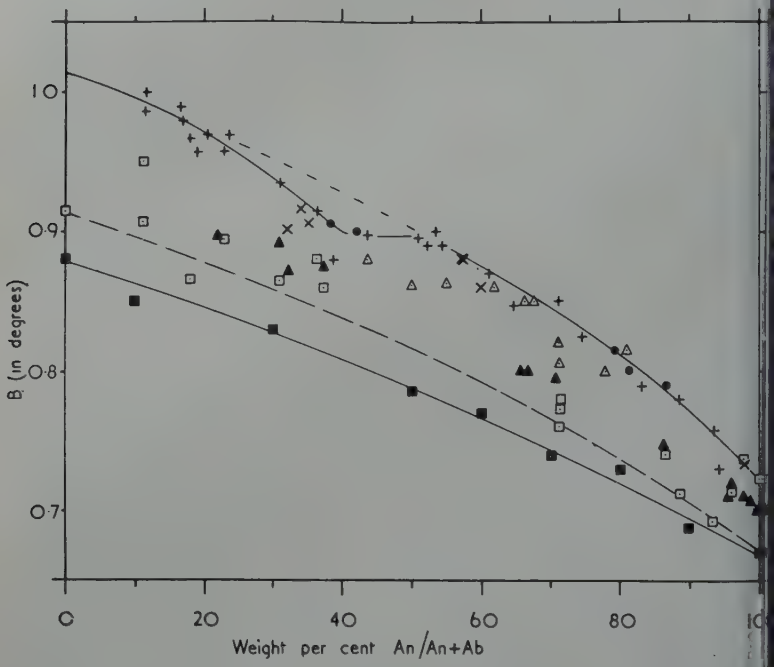

FIG. 2. The variation of B (2θ ($1\bar{1}1$)-2θ ($\bar{2}01$)) with wt. % An/(An+Ab). T symbols are the same as those in fig. 1. The significance of the curves is explai in the text.

angles given in part I) and An_{70} to An_{100} (to be discussed in detail late The high series is characterized by the values for quenched synthe plagioclases and the low series by the curve *ABCD* in fig. 1 which pas through those values of Γ with the greatest deviation from correspondi points on the high curve. (Obviously the discovery of plagioclase spe mens showing greater displacements than those plotted in fig. 1 wo result in a modification of the shape of the low curve.)

The high curve has the form of a gentle bow. The low curve ha curious shape, consisting of two linear portions of slightly different sl

ined by a short horizontal portion. The B curve for low specimens has milar characteristics, the central portion occurring in both cases at the me composition range, An_{40} to An_{50}. If the portion *CD* in fig. 1 is trapolated to An_0, it coincides within experimental error with the oint *A* for low albite. This and two other arguments to be considered the next few paragraphs lead to the suggestion that the low series as reviously defined may be metastable over the region An_0 to An_{50} and hat plagioclases stable at low temperatures in the strict thermodynamic ense would give Γ values along the line *AECD*.

The first argument is based on the single-crystal studies of Gay. he intermediate structure was found to extend from An_{20} to An_{70} Gay, 1956) and it would not be unreasonable to expect a linear or near near relation for the Γ and B values of fully ordered plagioclases. Gay nd Bown (1956) found that the plutonic specimens from An_{45} to An_{70} ave sharp subsidiary reflections which under drastic heat treatment ecame diffuse and gradually disappeared. Volcanic and most hypabys- l specimens yielded either no subsidiary reflections or only weak diffuse nes. In the composition range An_{20} to An_{41} even the plutonic specimens ave diffuse subsidiary reflections.

The correlation between the deviations from linearity of Γ and the ffuseness of the reflections from plutonic specimens is surely not rtuitous. The diffuseness might result from one or both of two causes: he structure might be partly disordered, for it has been shown that ansition to the disordered high-albite type of structure results in a ding and spreading of the subsidiary reflections; or the crystals might onsist of separate fully ordered regions that are too small to give sharp -ray reflections. It is known that the first possibility gives a change Γ: it is probable that the second possibility would not, though a liable assessment must await a full interpretation of the structural anges that occur in the intermediate region.

The second argument is based on the variation of Γ with the petro- gical environment. If equilibrium were easily attained, the ordered ructure would be formed even under hypabyssal and most volcanic nditions: only plagioclases from glassy rocks would give intermediate lues. Around An_{40}, a fairly continuous spread of values is found; in rticular, two specimens from the Southern Californian batholith viate rather more from the high curve than do plagioclases of similar mposition from the Skaergaard intrusion. Consequently it would not surprising if equilibrium had not been reached even in such a slowly oled complex as the Californian batholith. Smith (J. R.) and Yoder

grouped the plagioclases from the Skaergaard intrusion with those fro the Bushveld and Stillwater igneous complexes and focused attention o them because, 'All these plagioclases probably have similar therma histories, inasmuch as they all crystallized at basaltic magma tempera tures, and, as a result of the tremendous thickness of the intrusion cooled very slowly during and after crystallization. . . . Since thes plagioclases are the most slowly cooled of all magmatic plagioclases i the composition range which they cover (An_{36} to An_{86}) and since the make up a well-defined series by the criterion of reflection separation . . .' While the present authors agree that the Bushveld, Stillwater, an Skaergaard bodies are of tremendous thickness, they nevertheless believ that plagioclases from bodies such as the Southern California and th Nevada batholiths may well have had a better chance of approachin equilibrium. In addition, X-ray and optical evidence has been obtaine by Gay and Muir (to be published) that shows that many plagioclase from the Skaergaard intrusion have disordered structures. Consequentl the present authors see no reason for attaching especial importance t specimens from the Bushveld, Stillwater, and Skaergaard bodies a indicators of the low structural state.

Nevertheless, it is apparent that there is no conclusive evidence i favour of the hypothesis. Conclusive evidence might arise from eith of two sources. Either plagioclases that have undergone even longe 'annealing' periods may be found, or a theoretical proof might emerg when the nature of the ionic movements involved in the high–low i version has been fully elucidated. The small difference in Γ between th Skaergaard and other plutonic specimens indicates that very long perio at moderate to low temperatures will be required to produce the 'hypo thetical-low' specimens. The necessary conditions might be found i some metamorphic rocks but the situation might be complicated by th opportunity for mineralogical reconstitution with ensuing change o composition. For the present, however, such speculation must be di carded and the low series of plagioclases be regarded as defined by th curve *ABCD* (fig. 1) from An_{20} to An_{70}, and by the angles defined i paper I from An_0 to An_{20}.

Over the remaining composition range, An_{70} to An_{100}, there are man complex features. The difference in Γ values between synthetic an natural specimens decreases from 0·25° at An_{70} to about 0·06° for an orthites. Values of B occupy a band whose width ranges from 0·10° a An_{70} to 0·05° at An_{100}. The variation of both B and Γ with structura state is rather small and anomalies arise when attempts are made t

correlate the values of these quantities with the probable thermal history. Goodyear and Duffin found that from An_{80} to An_{100} there was little variation with thermal state of those angular separations dependent mainly on γ^*, and hence proposed that the An-content might be deduced from measurements of such angles. The present results, which are based on many more specimens, lead to more pessimistic results. Specimens differing in composition by as much as 7 % An give indistinguishable values of Γ; a determinative method of such low accuracy is of little practical application. The Γ values of quenched dry synthetic plagioclases give a more consistent variation with An-content than natural plagioclases and for this type of specimen the An-content from An_{80} to An_{100} can probably be estimated with an accuracy of 2–4 %.

There are striking anomalies in the values of Γ produced by heating. Before heating, Γ for a plutonic specimen of composition An_{88} lay on the curve that represents the average position for plutonic specimens; after heating Γ had increased by 0·09° and then lay 0·05° on the wrong side of the synthetic curve. Before heating, Γ for another plutonic specimen of composition An_{87} lay on the best curve for plutonic specimens but after heating Γ had decreased by 0·04°.

At first sight the values of B appear more promising. The quenched synthetic specimens give a continuous curve within experimental error. All the plutonic specimens except one give B values that lie on a continuous curve passing through the value for annealed synthetic anorthite. Volcanic specimens lie about midway between the two curves and the hypabyssal specimens lie close to the plutonic curve. Upon heating at elevated temperatures, the B values of the natural specimens from An_{70} to An_{96} move towards the synthetic curve. One volcanic specimen, of composition An_{98}, was annealed into the low-temperature state and it gives a B value on the plutonic curve. There is an excellent correlation between B and the structural state determined from single-crystal studies or deduced from the petrological environment except for the single plutonic specimen mentioned above. This specimen (SLR 354, An_{94}) comes from a calcic gabbro in the Southern California batholith and would be expected to give a B value on the plutonic curve; the deviation from the curve is only 0·02° but sufficient extra measurements were made to lower the random error of measurement to 0·005°. It would be surprising if this specimen were not in a fully ordered structural state; yet the B value indicates a state of inversion one-third of the way from that of other plutonic specimens.

The single-crystal studies of Gay and of Laves and Goldsmith have

shown that the structural changes in bytownites and anorthites are very involved and further unpublished work upon sodic bytownites has revealed additional complexities. In the intermediate plagioclases only one structural change occurs and a smooth variation of Γ is to be expected and indeed seems to exist. In the bytownites and anorthites there may be two interrelated structural changes between the low and the high forms. The transition between primitive and body-centred anorthite is dependent on the Na and Ca ions, and the change from body-centred anorthite to the high-albite-type structure depends on Al–Si order-disorder. As the velocity of the first structural transition is probably strongly dependent on the Al–Si order the structural arrangement in natural basic plagioclases may depend critically upon the cooling history of the felspar, and the simple classification that has been adopted here may not be valid.

From the present evidence, it is clear that in the composition range An_{70} to An_{100}, values of Γ are of little significance for the deduction of either structural state or composition. Apart from the single anomalous specimen, the values of B suggest that a good estimate of structural state may be made although the total variation is only five to ten times the experimental error. Although the anomalous specimen may have crystallized under singular conditions, the effect of factors other than structural state (iron or potassium atoms within the plagioclase lattice) may be important. The interpretation of structural state from values of B must be regarded, therefore, with some uncertainty, and its use is suggested only because optical measurements are particularly insensitive in this region and because it avoids the laborious techniques of single-crystal methods. It is to be noted that the peaks *F* and *G* of Goodyear and Duffin are thought to be dependent on the same dimensional variables that the B plot employed; apart from any difficulty in the accurate location of peak *F* mentioned before, the separation *F*–*G* must be regarded as subject to all the limitations described above for the B plot.

Practical applications of the powder methods.

The authors feel that the non-specialist, faced with the large volume of original work that has appeared in recent years, may find it laborious to decide how the various techniques are to be most efficiently applied, so in this section they attempt to assess the relative merits of the various techniques in the estimation of An-content and structural state.

The An-content may be determined most reliably by the standard analytical methods or may be estimated by spectroscopic methods; both

e destructive and give no information about the structural state of the agioclase. In routine petrographic studies, optical methods are in neral use. The An-content may be estimated by comparing the re-active indices with the standard curves that have been obtained by eraging over numerous analysed plagioclases. The variation of re-active indices with structural state has not been systematically studied, though Kano (1955) and Schwarzmann (1956) have recently shown that e standard curves of Chayes (1952), while applicable to low plagio-ases, may lead to serious error (5 % An) for those sodic plagioclases at have a disordered structure. The structural state appears to have tle effect on the refractive indices of calcic plagioclases. The optic ial angle varies considerably with structural state for albites and igoclases, though the variation over the remaining composition range small.

The relative orientation of the crystallographic and optical axes is of nsiderable value, for under favourable conditions it provides an esti-ate of both the An-content and the structural state. A large body of ta has been obtained by universal-stage methods (see van der Kaaden, 51, for example) and from this the An-content may be estimated with theoretical precision of about 2 % if the experimental error is 2°. The ta on the effect of structural state are less complete, but indicate that ght stages in the transition from low- to high-albite might be dis-nguished: for intermediate specimens, the separable stages fall to ree, whilst for basic specimens (for which few observations are avail-le) the determination of structural state appears to be impracticable.

Although a great wealth of experimental information on the optical operties of plagioclase felspars has been obtained in the past, there has en little correlation between the results of one worker and another: mprehensive studies of the effects of strong heating and of the petro-gical environment have not so far been attempted. The results of any workers have shown a wide spread of optical parameters for ystals taken from the same hand specimen. While some of the spread undoubtedly caused by variable An-content and structural state, her factors such as poor definition of crystallographic elements and lid-solution of impurity atoms may well be of importance. The authors ve found from their X-ray work that the more comprehensive the study, e greater the number of exceptions and variations that are found, d they would expect a similar situation for optical studies. Conse-ently, while they believe that the broad outlines of plagioclase optics ve been established, they would urge that detailed interpretation

be made with some caution until the optical variations have bee authoritatively established by wide-ranging complete studies. Subjec to this limitation, the present value of optical methods may be sum marized: They are rapid and non-destructive. The composition an structural state of albites and oligoclases may be reliably estimate from the optical orientation with confirmation, if desired, from th refractive indices and optic axial angle. For andesines and labradorites the An-content may be reliably estimated from the refractive indices o from the optical orientation; the structural state can only be estimate in a crude manner from the optical orientation. For bytownites an anorthites, although the An-content may be determined either from th refractive indices or the optical orientation, there is little hope o deducing the structural state from the optical properties.

Turning to the X-ray powder methods, the reflection angles a governed by six fundamental variables, the three lattice angles and th three lattice repeats. Unfortunately in sodic plagioclases b^*, c^*, α^*, an γ^* vary in a similar manner (but γ^* much more than the others); α appears to be unreliable, probably because of solid-solution of potas felspar, whilst β^*, although showing an independent form of variatio changes so little that it is not of practical value. Hence there is effe tively only one useful form of variation, so that either the structural sta or An-content must be known before the other can be estimated. I practice, the cell dimensions themselves are not used as variables an some simple angular relationship between the X-ray reflections measured. As discussed earlier, the Γ function is most useful from An to An_{70}, whilst the three angles in part I are to be preferred from A to An_{20}. The method permits the detection of as many as fifty ste between high- and low-albite. The sensitivity falls rapidly to An and then more slowly to An_{70} where several stages of structural variatio may still be recognized. From An_{70} to An_{100}, two variable paramete may be detected in the angular positions of the powder diagrams, bu their variations are small, and factors other than An-content and stru tural state appear to have significant effects. However, an estimate structural state may be made from the B variation, and appears to l reliable for the majority of specimens. Since the completion of the pr sent study, Laves and Goldsmith (1956) have demonstrated that t powder pattern of anorthite contains a reflection whose intensity provid a measure of the transition from the primitive to the body-centr structure. It is hoped that further study of the variation of this refle tion with An-content and structural state will lead to an accurate dete

minative method—especially as the other methods are so ineffective in his composition range.

Single-crystal X-ray methods can yield an unambiguous estimate of structural state for intermediate and basic plagioclases from An_{40} to An_{100}. An elegant method for the independent determination of An-content in the intermediate region (An_{40}–An_{70}) has been described. Nevertheless, it is unlikely that routine determinative methods can be developed; for apart from the inherent technical difficulties, most of the methods of estimating structural state involve the observation of the intensity, and perhaps diffuse character, of certain weak reflections. Thus, for plagioclase felspars, the chief value of single-crystal methods will be in the provision of a rigorous standard against which the simpler routine methods may be checked.

Finally, suggestions will be presented for the application of these methods to mineralogical and petrological studies. First, for routine survey work, optical measurements obtained from thin sections would appear to be sufficient for the characterization of plagioclase felspars. Secondly, in petrogenetic studies, where greater reliability is required, universal-stage measurements could be supplemented by powder X-ray measurements and the determination of refractive indices. The region An_{70} to An_{100} is particularly difficult, for the estimates of structural state, obtained from the powder X-ray measurements, are not definitive. Thirdly, if a complete description of the plagioclase felspar is required, all the methods may be employed with advantage. The An-content may be obtained from chemical analysis and checked by the refractive indices. The structural state may be determined by both powder and single-crystal X-ray methods, the former being more valuable for sodic specimens and the latter for calcic ones. The An-content and structural state can then be checked by a determination of the optical orientation.

Although this threefold scheme is of wide application, the peculiarities of the various methods may be advantageously employed to suit the special requirements of a particular problem. Many uncertainties still remain, particularly in the knowledge of the optical properties, but it is hoped, and indeed expected, that their clarification will not enforce serious modification of these conclusions.

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Rare earths in some niobate-tantalates.

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Summary. Rare-earth distribution has been determined for fourteen niobate-antalates containing essential rare earths. Those minerals with more than about 5 % TiO_2 (and corresponding to either priorites or members of the euxenite–olycrase series) have Yt as the dominant rare earth with the heavier lanthanons[1] n excess of the lighter lanthanons. Those minerals with less than about 4 % TiO_2 nd less than about 30 % $(Yt,Ln)_2O_3$ (and corresponding to samarskites) also ave Yt as the dominant rare earth but they show a marked concentration of d + Tb + Dy over the other lanthanons. It is tentatively suggested that this power f selective lanthanon enrichment may be characteristic of samarskite among the iobate-tantalates examined.

THE identification of those metamict minerals containing essential Nb, Ta, and rare earths is not a straightforward matter. Crystal morphology is the most valuable single diagnostic feature, but frequently he minerals have poorly developed faces or occur massive. Thus it is hat in the field it is not always possible to distinguish between mineral roups, and consequently laboratory data assume considerable impor-ance. Both X-ray and differential thermal techniques have been em-loyed (e.g. Berman, 1955; Kerr and Holland, 1951) but the methods lo not always give clear-cut results. Chemical data are certainly more seful and, as will be shown, division into certain mineral series or roups can be made on such data alone. Nevertheless, the rare-earth iobate-tantalates have not been extensively analysed, partly, no doubt, ecause of the difficulties they present to the analyst. Estimation of Nb and Ta has always required considerable skill using the classical nethods, and the modern methods are not devoid of problems especially or minerals rich in Ti.[2] Data on the individual rare earths are noticeably carcer than data on Nb and Ta and it seemed worth while carrying out small-scale survey of readily available minerals like samarskite and nembers of the euxenite–polycrase series to determine if and in what

[1] Lanthanon, and the symbol Ln, is a general term for all the rare-earth elements rom La to Lu; Sc and Yt are not included.

[2] The method of Williams (1952) seems to need modification for minerals like uxenite.

respects distribution of the rare earths varied in the different minerals. Members of the rare earth group are taken to consist of Sc (atomic number $Z = 21$), Yt ($Z = 39$), and the lanthanons, with atomic numbers $Z = 57$ to $Z = 71$ inclusive; Pm ($Z = 61$) is sensibly absent in nature.

Identification of the minerals.

The more common rare-earth niobate-tantalates can be conveniently divided into two major groups on the basis of Ti content:

Those with *Ti exceeding about 15 % TiO_2* include polymignite, members of the euxenite–polycrase series, and members of the aeschynite–priorite series. Few analyses of polymignite are available but the mineral is here unique in containing essential Zr. Sufficient analyses of the other minerals are available to convince both casual and careful observers of the chemical complexities of the series; unfortunately the basis for division in the euxenite–polycrase series revolves round the Nb, Ta, and Ti contents whereas that for division in the aeschynite–priorite series depends on the rare-earth (and Th) content. In euxenite the atomic ratio Ti/(Nb+Ta) is less than 3 whereas in polycrase it exceeds 4 (Brøgger, 1906); contents of TiO_2 in the series range from about 15 to 35 wt. %; usually Nb exceeds Ta although the ratio Nb/Ta can vary at least by a factor of ten. The name tanteuxenite has been proposed for minerals otherwise euxenite when Ta_2O_5 exceeds Nb_2O_5 on a wt. % basis; no new name has been proposed for Ta-rich polycrase. The description tantalian euxenite or tantalian polycrase would seem adequate to imply knowledge of Nb, Ta, and Ti contents in the mineral. In aeschynite the lighter lanthanons (La, Ce, &c.) are the dominant constituents of the rare earths whereas in priorite Yt and the heavier lanthanons (Gd, Dy, &c.) are dominant; further, in aeschynite Th is higher (e.g. 15 % ThO_2) than it often is in priorite (e.g. 8 % ThO_2). In the series TiO_2 varies from about 20 to 35 % and although the extent of the variation in the ratio Nb/Ta is not well known no Ta-rich varieties have been described.

It must be appreciated that a partial chemical analysis for the elements or chemical groups Nb and Ta, Ti, and Yt and heavier lanthanons will not enable a distinction to be made between priorite and a member of the euxenite–polycrase series. Knowledge of the U and Th contents is, however, useful in so far as U can be and often is higher in members of the euxenite–polycrase series than in priorite; moreover, Th can be and often is higher in priorite than in members of the euxenite–

olycrase series; and the atomic ratio U/Th is generally higher in mem-ers of the euxenite–polycrase series than in priorite.

The minerals described in this paper as members of the euxenite–olycrase series all have fairly low Th contents (table I, cols. 1–10) and he U/Th ratios at least exceed unity. X-ray diffraction patterns of the pecimens after heating in air at 700° C. for 6 hours (Berman, 1955) can e divided into three types. Those of minerals nos. 1, 2, 3, 6, and 10 able I) gave comparable patterns similar to that for (heated) lyn-ochite, a rare-earth niobate-tantalate rich in the lighter lanthanons Butler, 1957*c*); those of minerals nos. 4, 5, 7, and 8 gave comparable atterns similar to that of samarskite from North Carolina (no. 12) after he latter had been heated in air at 1000° C. for $2\frac{1}{2}$ hours; mineral no. gave a unique pattern.

Those rare-earth niobate-tantalates with *Ti below about 4 %* TiO_2 nclude the fergusonite–formanite series, yttrotantalite, and samarskite. he basis for division in the fergusonite–formanite series is the Nb/Ta tomic) ratio which exceeds unity for fergusonite and is below unity for rmanite; rare earths nearly always total above 30 % and average earer 40 % $(Yt,Ln)_2O_3$. Samarskite usually contains between 10 and 5 % rare earths; Nb is dominant over Ta. Both U and total Fe usually xceed 4 %, whereas some members of the fergusonite–formanite series an be low in both these constituents. Yttrotantalite is chemically milar to samarskite except that Ta dominates over Nb.

The minerals described in this paper as samarskite have rare earths talling between 12 and 25 % $(Yt,Ln)_2O_3$ (table I, cols. 11–14); semi-uantitative tests showed that the Nb/Ta ratio was appreciably greater an unity. X-ray diffraction patterns of the specimens nos. 11, 12, and 4 after heating in air at 1000° C. for $2\frac{1}{2}$ hours were similar (and com-arable with those of specimens nos. 4, 5, 7, and 8 after they had been eated in air at 700° C. for 6 hours); that for specimen no. 13 gave a nique pattern.

Analytical methods.

Rare earths plus thorium were determined via the fluorides, perchlor-es, and oxalates using a modified method after Gordon *et al.* (1949) escribed elsewhere (Butler, 1957*b*). The separation is probably not itable for the quantitative estimation of Sc as its oxalate is not com-etely insoluble under the chosen conditions of oxalate precipitation; owever, absence of Sc in the minerals (i.e. not in the separated oxides) as confirmed spectrographically (sensitivity about 0·07 % Sc_2O_3).

TABLE I. Analytical data for some Nb–Ta–Yt–Ln minerals. Nos. 1–10 are thought to be either priorites or members of the euxenite-polycrase series; nos. 11–14 are thought to be samarskites. Individual rare earths were determined spectrographically in and reported on the basis of chemically separated $(Yt,Ln)_2O_3+ThO_2$. (Sc_2O_3 and Eu_2O_3 below 0·1 % throughout, La_2O_3 below 0·1 %, Pr_6O_{11} below 0·3 %, and Lu_2O_3 below 0·1 % where indicated by —. No data for Ho_2O_3.) Total rare-earth oxides of the formula shown, TiO_2, and ThO_2 were determined chemically on a mineral basis. R^{3+}, ionic radii (Å.) of trivalent rare-earth cations. δ, percentage standard deviations of estimate of individual rare earths from duplicate spectrograms; the higher values for La_2O_3 and Pr_6O_{11} are for lower concentration estimates.

	R^{3+}.	1.	2.	3.	4.	5.	6.	7.	8.	9.	10.	11.	12.	13.	14.	δ.
Yt_2O_3	0·92	59·2	59·5	47·0	46·8	47·0	39·7	49·8	45·8	44·4	46·0	44·7	44·0	49·2	58·6	5
La_2O_3	1·14	0·10	—	0·15	0·12	—	0·12	0·40	0·35	0·10	0·38	—	—	—	0·13	7–10
CeO_2	1·07	0·52	0·60	1·47	0·96	0·57	1·10	1·25	2·79	1·45	2·48	0·92	1·10	1·28	0·89	7
Pr_6O_{11}	1·06	—	—	0·63	0·56	—	0·65	0·53	1·12	0·66	0·78	0·50	0·65	0·76	—	7–10
Nd_2O_3	1·04	0·93	0·85	3·74	2·08	1·37	3·01	1·98	7·90	3·60	5·75	2·58	3·84	3·46	1·97	5
Sm_2O_3	1·00	1·16	0·85	3·17	1·72	1·94	4·75	2·22	7·24	3·88	2·18	5·55	10·2	6·97	3·20	5
Gd_2O_3	0·97	4·04	3·30	6·95	5·06	6·70	9·64	6·26	11·7	9·77	3·26	16·9	27·6	12·8	10·9	6½
Tb_4O_7	0·93	1·31	1·02	1·59	1·25	1·45	2·24	1·54	2·56	2·23	0·81	3·70	4·90	3·40	2·93	6
Dy_2O_3	0·92	7·00	6·47	9·14	7·75	10·5	11·8	10·6	11·0	12·0	2·98	16·0	13·6	13·6	14·2	5
Er_2O_3	0·89	8·32	7·14	9·72	10·9	10·2	9·35	8·06	6·77	10·0	5·65	4·15	1·21	4·82	3·88	13
Tm_2O_3	0·87	0·84	1·10	1·00	1·37	1·13	0·93	0·93	0·58	0·97	1·06	0·26	—	0·52	0·33	10
Yb_2O_3	0·86	8·82	11·6	10·0	12·4	6·55	6·17	9·68	3·60	6·99	11·2	1·35	0·11	2·81	2·19	5
Lu_2O_3	0·85	0·83	1·30	0·96	1·69	1·70	0·76	0·83	0·50	0·84	3·75	0·25	—	0·46	0·39	30
ThO_2	—	6·18	1·29	11·20	7·78	10·10	9·27	5·20	12·80	n.d.	16·20	2·38	tr.	nil	nil	—
Sum	—	99·25	95·02	106·72	100·44	99·21	99·49	99·28	114·71	96·89)	102·48	99·24	107·21	100·08	99·61	—
$(Yt,Ln)_2O_3$		25·96	27·71	30·07	23·10	20·30	28·28	21·77	25·47	(20·85)	20·86	16·85	17·70	12·44	24·68	
TiO_2		25·0	24·3	29·6	24·9	20·5	24·4	27·0	20·0	25·0	21·8	2·10	2·17	1·60	1·20	
ThO_2		1·71	0·36	3·79	2·07	2·28	2·89	1·44	3·73	n.d.	4·04	0·41	tr.	nil	nil	

1. Skillnabo, Västergötland, Sweden.
2. Mattawan Township, Nipissing District, Ontario, Canada.
3. South West Africa.
4. Nanseke, Uganda.
5. Kjaelevann, Vegusdal, Norway.
6. Arendal, Norway.
7. Brazil.
8. Vaalputs portion of Nousees, 5 miles east of Nousees Homestead, South-west Africa.
9. Fitamalama, Madagascar.
10. Rio Ligonha, Moçambique.
11. Bjortjenn, Iveland, Norway.
12. Mitchell Co., North Carolina, United States. B.M. 68781.
13. Divino Ubá, Minas Gerais, Brazil. B.M. 1926,246.
14. Madras, India.

horium was determined by decomposing the HF-insoluble fluorides ith sodium bisulphate and precipitating thorium as iodate using the ethod of Meyer and Speter (1910); it was found advantageous to ice-ool the solution prior to addition of sodium sulphite and to allow the olution to become completely iodine-free (i.e. decolorized) prior to ddition of ammonium hydroxide. Titanium was determined colori-etrically using the yellow Ti-H_2O_2 compound; uranium was estimated pproximately using the method of Bowie (1949). Individual rare earths ere determined spectrographically on the chemically separated rare arth plus thorium oxides (Butler 1957*a*); it was not found possible to stimate Ho with adequate precision although it probably reaches 2 % Io_2O_3 for the samples rich in Dy and Er. The errors of determination ssociated with the spectrographically determined oxides are shown in able I (that for chemically determined Th is relatively negligible). The ummations of the individual oxides should be, say, $\frac{1}{2}$–2 % less than 100 ccording to the Ho_2O_3 content and it is possible to use this knowledge of he total to 'improve the data' for the individual oxides by adjusting the xide percentages in proportion to their variances (squares of actual eviations) using Merriman's method of least squares. For oxides from inerals 2, 3, and 8 with totals of 95, 107, and 115 respectively improved alues for oxide percentage estimates might be of use were it not for he probability that Ho_2O_3 is likely to be nearer, say, 2 or 3 % than $\frac{1}{2}$ %; ith or without improved estimates the rare-earth distribution trends e the same. For oxides from mineral 12, however, the total is 107·2, it Ho_2O_3 is absent (i.e. below $\frac{1}{2}$ %): improved estimates have been tained to give more precise data for this very gadolinium-rich mineral. Table II, col. 12, shows the atomic percentages of the lanthanons in e total lanthanons derived by Merriman's method from the data in ble I, col. 12. All other data in table II are derived directly from cor-sponding data in table I.

Previous work.

The determination of rare earths by classical chemical methods has rked limitations and often the best that can be accomplished is an imation of Ce and a division into the cerium group (La, Ce, Pr, Nd, l Sm), the terbium group (Eu, Gd, and Tb), and the yttrium group , Dy, Ho, Er, Tm, Yb, and Lu) (Hillebrand *et al.*, 1953). Neverthe-, several analysts were bold enough to publish figures for, specifically, O_3 half a century ago, before, indeed, the exact number of elements

TABLE II. Atomic percentages of lanthanons in total lanthanons excluding Ho. For localities see Table I. (*Z*, Atomic number.)

	Z.	1.	2.	3.	4.	5.	6.	7.	8.	9.	10.	11.	12.	13.	14.
La	57	0·34	—	0·35	0·31	—	0·27	1·03	0·70	0·22	1·07	—	—	—	0·31
Ce	58	1·68	1·94	3·28	2·29	1·47	2·34	3·06	5·24	2·96	6·62	1·88	1·82	2·67	2·32
Pr	59	—	—	1·42	1·35	—	1·40	1·31	2·12	1·37	2·11	1·03	1·09	1·60	—
Nd	60	3·07	2·81	8·54	5·09	3·60	6·55	4·97	15·2	7·55	15·7	5·40	6·53	7·39	5·25
Sm	62	3·70	2·71	6·96	4·05	4·95	10·0	5·40	13·4	7·85	5·79	11·2	16·7	14·4	8·25
Gd	64	12·4	10·1	14·7	11·5	16·3	19·4	14·6	20·9	19·0	8·31	32·8	43·5	25·3	27·0
Tb	65	3·98	3·03	3·26	2·74	3·42	4·40	3·50	4·43	4·19	1·99	6·95	7·46	6·52	7·05
Dy	66	21·4	19·3	18·8	17·1	24·9	23·1	23·9	19·1	22·7	7·37	30·0	20·8	26·2	34·1
Er	68	24·2	20·8	19·5	23·6	24·3	17·9	17·8	11·5	18·5	13·6	7·61	1·81	9·04	9·10
Tm	69	2·42	3·10	1·98	2·91	2·58	1·76	2·02	0·97	1·77	2·53	0·47	—	0·97	0·77
Yb	70	24·9	33·0	19·5	25·8	14·7	11·5	20·7	5·90	12·5	26·2	2·40	0·16	5·13	4·98
Lu	71	2·31	3·63	1·84	3·48	3·77	1·40	1·75	0·82	1·49	8·69	0·44	—	0·83	0·88

onstituting the lanthanon group was established. The appearance of heir results in subsequent and modern textbooks is no reason for not egarding the data as tentative or even highly approximate. The first vorker to produce acceptably accurate semi-quantitative data for ninerals like euxenite and samarskite was, of course, Goldschmidt. Together with Thomassen (Goldschmidt and Thomassen, 1924), he volved an X-ray spectrographic technique and showed these minerals o be poor in La, Ce, &c., but rich in Yt and the heavier lanthanons like Dy, Er, and Yb. Only one samarskite and six members of the euxenite–olycrase series were analysed and the semi-quantitative data did not uggest a distinction between the different minerals on a basis of rare-arth distribution. However, Goldschmidt demonstrated that any lan-hanon with an odd atomic number was invariably less abundant than hose lanthanons with adjacent (and even) atomic numbers; he also xplained the absence of Eu ($Z = 63$) as due to the ease of reduction of Eu^{3+} to Eu^{2+} and the consequent disassociation of the larger divalent on (radius 1·24 Å.) from the smaller trivalent lanthanon ions (varying rom 1·14 Å. for La^{3+} to 0·85 Å. for Lu^{3+}).

Bjørlykke (1935) used X-ray spectrographic methods to study rare arths and other elements, including Ti, occurring in eight samarskites nd seventeen different members of the euxenite–polycrase series. He oncluded that '. . . the distribution of the rare earth elements [in the amarskites] is very like that of the euxenites. . . '. However, in his list f estimated intensities of K_α and L_α lines obtained in X-ray spectro-rams of the minerals it is noticeable that the Yb line intensity is below on an arbitrary scale 0–10 for 6 of the 8 samarskites and above 1 for 2 of the 17 members of the euxenite-polycrase series; further, the Dy ne is the most intense lanthanon line in 7 of the samarskites. He con-med that all 25 minerals were poor in the lighter lanthanons La, Ce, r, and Nd.

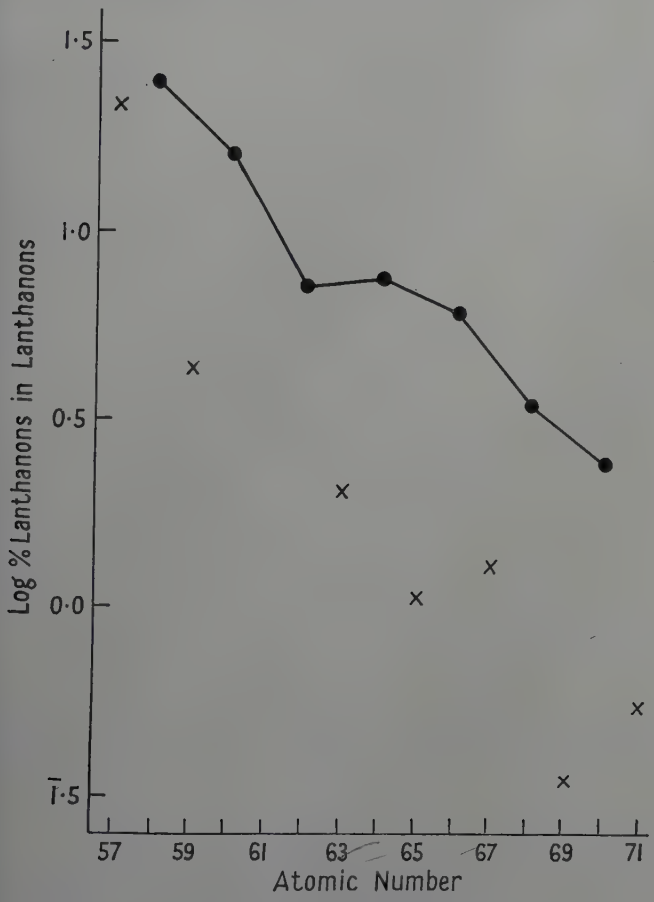
Using a refined X-ray spectrographic technique Sahama and Vähätalo 939) were able to produce percentage concentration figures for in-vidual rare earths in several niobate-tantalates. The materials re-rred to by the authors as samples of wiikite were ill-defined mixtures alteration products of pegmatitic minerals like euxenite or samarskite, nd were evidently capable of very wide compositional variations (Lokka, 28). Nevertheless, all the wiikites were consistent in having Yt as sily the most abundant rare earth, and Yt and the heavy lanthanons l, Tb, Dy, Ho, Er, Tm, Yb, and Lu were from 2·5 to 19 times as undant as the other lanthanons on an atomic basis.

Discussion.

Cols. 1–10 of table I show data for ten niobate-tantalates rich in TiO_2 from widespread localities where Nb is thought or known to dominate over Ta. The total rare-earth percentages vary only between 20 and 30 and, as expected, do not correlate with either ThO_2 or TiO_2 content. Yt is easily the most abundant rare earth in all the samples but, again, cannot be correlated with total rare-earth content. On an atomic basis Yt represents between about 55 (nos. 6 and 8) and 75 (nos. 1 and 2) % of the rare earths and invariably the heavier lanthanons, viz Gd, Tb, Dy, (Ho), Er, Tm, Yb, and Lu, substantially exceed the lighter lanthanons. Lanthanon distribution in the minerals is thus markedly different from that obtaining in the lithosphere (fig. 1) where the average abundance of the *Z*-even lanthanons falls fairly regularly from Ce to Yb by a factor of very approximately 10 and those of the rarer *Z*-odd lanthanons fall from La to Lu by a factor probably exceeding 10 (excluding the highly unstable Pm, $Z = 61$). It is possible that one or more of the minerals crystallized from an environment impoverished in the lighter lanthanons (relative to the average lithosphere abundance of the lanthanons) by previous crystallization of, say, the most common rare-earth mineral monazite, which contains the light lanthanons almost to the exclusion of Yt and the heavy lanthanons (Murata *et al.*, 1953). This type of pre-enrichment can hardly have obtained for all the 10 minerals here analysed and it may be concluded that they are of the type described by Goldschmidt and Thomassen as showing a selective rare-earth assemblage. Nevertheless, the degree of selectivity is not as high as in, say, thalenite or thortveitite, and it will be seen from table II that minerals nos. 1–9 each contain at least 3 lanthanons exceeding 15 at. % of the total lanthanons (omitting Ho): there are accordingly no marked peak concentrations of individual lanthanons. Moreover, the lanthanons vary appreciably from specimen to specimen; Ce, for instance, varies by a factor of $3\frac{1}{2}$ from 1·4 to 5·2 at. % in the lanthanons and Yb varies by a factor of $6\frac{1}{2}$ from 5·9 to 33 at. % on the same basis. Dy, however, is much less variable (17·1 to 24·4 at. % in minerals 1–9 and 7·3 at. % in mineral 10) and it is quite probably the optimum-sized lanthanon for the euxenite (or priorite) structure; Yt^{3+} and Dy^{3+} have identical radii.

In a plot of lanthanon concentration against atomic number three trends are discernible. Thus the minerals from Västergötland, Sweden (no. 1), Ontario, Canada (no. 2 and fig. 2), South-west Africa (no. 3) and Nanseke, Uganda (no. 4), are comparable in showing a more or less regular increase in concentration for *Z*-even lanthanons with increasing

atomic number. Er^{3+} and Yb^{3+} are, of course, slightly smaller than Dy^{3+} and their presence in excess of Dy does not impose much strain on the crystal structure, more especially as more than half of the rare-earth positions in the lattice are occupied by Yt^{3+}. It is not, however, possible



G. 1. Lanthanon distribution in 'average meteorites' after Suess and Urey (1956). Points representing the *Z*-even lanthanons are joined.

decide if the relative concentration of Er and Yb compared with that the other minerals was due to rate of crystallization or temperature of rystallization of the mineral rather than a relative concentration of Er d Yb in the environment in which the minerals crystallized. The odd lanthanons also reflect the concentration of the smaller-sized

lanthanons in minerals nos. 1–4. Tb always exceeds Pr which in turn exceeds La but there is not invariably an increase in concentration of either Tm or Lu over Tb. There is, however, an appreciable increase in the ratio Tm/Tb over that obtaining in the lithosphere (fig. 1). The irregular trend of Lu/Tm or Lu/Yb ratios is possibly emphasized by the

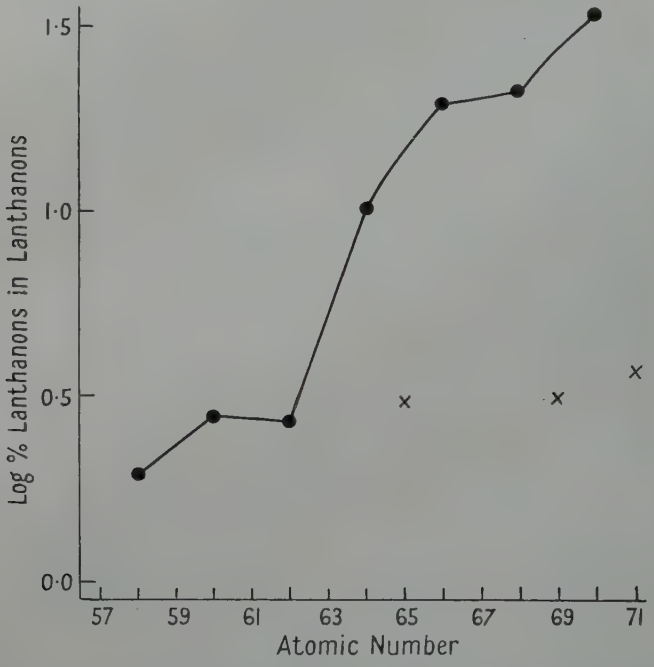


Fig. 2. Lanthanon distribution in Ti-rich niobate-tantalate, Mattawan Township, Nipissing District, Ontario, Canada (no. 2). Points representing the Z-even lanthanons are joined.

high standard deviation associated with the determination of Lu; however, it is also likely that it indicates that there is no tight distinction in lanthanon distribution between these four minerals and those in the following groupings.

The minerals from Vegusdal, Norway (no. 5), Arendal, Norway (no. 6), Brazil (no. 7), Nousees, South-west Africa (no. 8), and Fitamalame, Madagascar (no. 9), show, like those in the first grouping, about 20 at. % Dy and about 3–4 at. % Tb in their lanthanons but the concentrations of Er and Yb and of Tm and Lu tend to fall from that of Dy and Tb respectively (fig. 3, specimen no. 6).

Their higher Gd content is a consequence of the limited range in both otal rare-earth content of the mineral and the Yt content in the rare arths. These four minerals are the only ones with Gd, Tb, and Dy

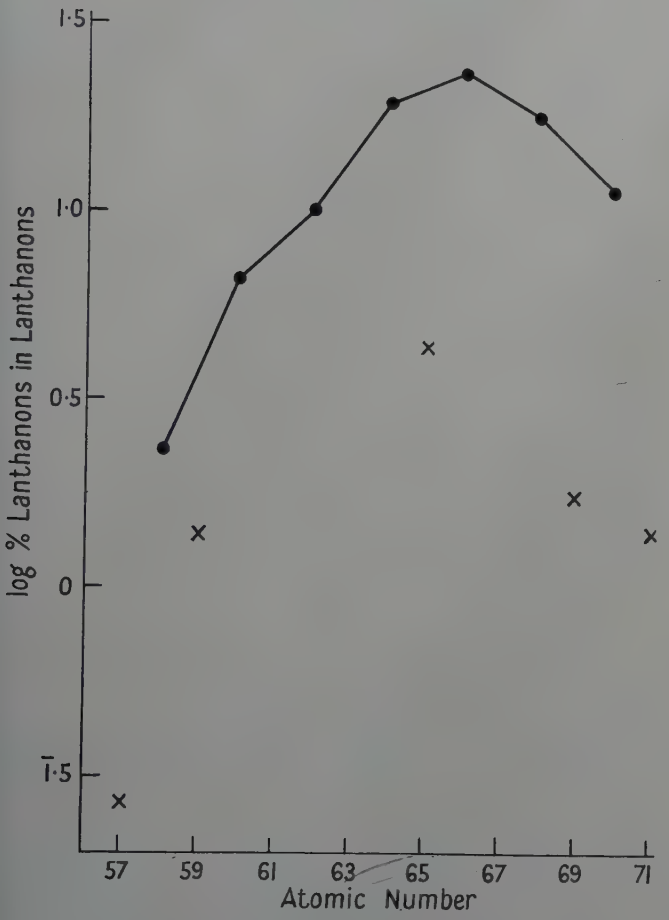


FIG. 3. Lanthanon distribution in Ti-rich niobate-tantalate, Arendal, Norway (no. 6). Points representing the *Z*-even lanthanons are joined.

talling above 40 at. % of the lanthanons and in the mineral from ousees, South-west Africa, Gd exceeds Dy, although by rather less an in the lithosphere. The tolerance of these minerals to lanthanons th ionic sizes above that of Dy^{3+} and Yt^{3+} is well seen in the latter neral where Nd+Pr+Ce exceeds 22 at. % in the lanthanons; the

weak concentrating factor of the mineral for a particular sized lanthanon was insufficient to offset the relatively high concentration of the lighter lanthanons in the environment from which the mineral crystallized.

The mineral from Rio Ligonha, Moçambique, shows a lanthanon distribution unique for the Ti-rich niobate-tantalates here investigated

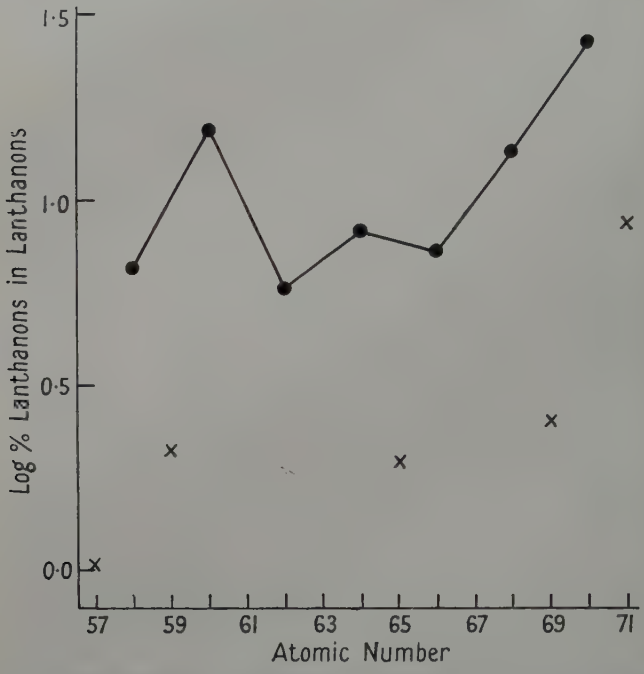


FIG. 4. Lanthanon distribution in Ti-rich-niobate tantalate, Rio Ligonha, Moçambique (no. 10). Points representing the Z-even lanthanons are joined.

(fig. 4). There are peak concentrations for Nd ($Z = 60$) and Yb ($Z = 70$) among the Z-even lanthanons and for Lu ($Z = 71$) among the Z-odd lanthanons; Pr ($Z = 59$) and Tb ($Z = 65$) show similar concentrations. This concentration-distribution type has been observed in a more severe form in davidite by Dixon and Wylie (1951). It is possible that the smallest lanthanons occupied sites in the mineral structurally dissimilar to those occupied by the largest lanthanons; this is difficult to prove for a metamict mineral. An alternative explanation is that the middle-sized lanthanons, especially Gd, Tb, and Dy, were relatively impoverished in the fluid from which the mineral formed by the previous (or contemporaneous) crystallization of another mineral strongly concentrating

st those lanthanons; samarskite would fill this role admirably. The gh Lu content in the Moçambique mineral (0·93 % Lu_2O_3) and the atively high Lu/Yb ratio is of considerable interest in view of the k of Sc (brief mention of a similar state of affairs in a sample of gusonite has been made by Wylie (1954)). Lu^{3+} and Sc^{3+} have ionic es 0·81 Å. and 0·85 Å. respectively and diadochy is clearly possible for e pair of elements; on size consideration alone it could be supposed at Sc would have little difficulty in substituting for Lu in minerals rich Lu and the heaviest lanthanons. However, Sc breaks away from Lu an early stage in magmatic differentiation and is a well-noted trace ment in the common rock-forming ferromagnesian minerals, due to e substitution of Sc^{3+} (0·81 Å.) for Fe^{2+} (0·74 Å.). Presumably the pture of much of the larger Lu^{3+} (0·85 Å.) by Fe^{2+} would impose an cessive strain in the lattice of ferromagnesian minerals, but data on in such minerals are lacking. This size strain may be a reason why e electronegativity approach suggested by Ringwood (1955) does not e apply.

The electronegativity (E) of Lu at 1·05 volts is 0·25 volts less than that Sc; according to Ringwood this should favour substitution of Lu er Sc) for a suitably sized ion (where diadochy between all 3 ions would possible). The difference in behaviour between Sc and Lu may more nveniently be correlated with their differences in ionization poten-ls; the ratio [Ionization potential $Sc(Sc^{2+} \rightarrow Sc^{3+})$]/[Ionization poten-l $Lu(Lu^{2+} \rightarrow Lu^{3+})$] is 1·29 which, according to Ahrens (1952 and 53) is rather high for close geochemical coherence to be expected. This newhat weak coherence of Sc and Lu is in contrast to that for the gest trivalent rare earths La and Ce even though their size difference pressed as a percentage of the mean exceeds that of Sc and Lu (6·4 % inst 4·8 %). The ionization potentials of La and Ce are closely ilar and geochemical coherence is further helped by the absence of y common lithophile element with ionic radius near to or just a little ger than that of La. Only Hg^{2+} (1·10 Å.), Sr^{2+} (1·12 Å.), and Pb^{2+} 20 Å.) have radii between 1·07 Å. and 1·22 Å.

It has been stressed that the minerals nos. 1–10 need not be members the euxenite–polycrase series on the basis of U and Th contents and ios; they may be members of the priorite–aeschynite series albeit r the priorite end. If the mineral from Nousees, South-west Africa, onsidered as a member of the latter series then its high atomic pro-tion of Yt and the heavier lanthanons compared with the lighter thanons would stamp it as being very near the theoretical priorite

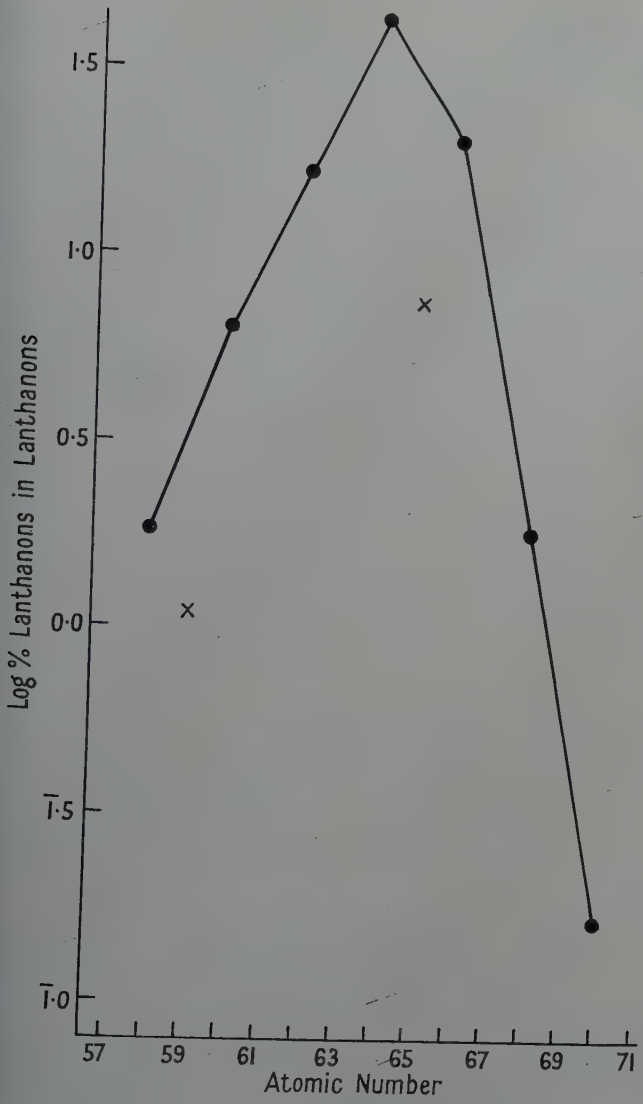
end member of the aeschynite–priorite series; the light lanthanons La–Sm inclusive account for only 16 at. % of the total rare earths.

Data for these minerals—in particular those showing the restricted abundance of the light lanthanons—formed a basis for suggesting that lyndochite, with 55 at. % of light lanthanons constituting its rare earths, was chemically more comparable to a mid-compositional member of the aeschynite–priorite series than to members of the euxenite–polycrase series (Butler, 1957*c*). Nevertheless, it is to be appreciated that this survey is a preliminary and small-scale one; minerals morphologically like euxenite and compositionally similar to lyndochite are not necessarily rare.

Cols. 11–14, table I, show data for four niobate-tantalates poor in TiO_2, with Nb exceeding Ta, and with ThO_2 below 1 %. The total rare-earth contents vary from about 12 to 25 at. % and these data are not inconsistent with all four minerals being samarskite. Yt is easily the dominant rare earth and its percentage range (in total rare earths) is like that in the Ti-rich niobate-tantalates previously discussed. Yt accounts for 53 at. % of the rare earths in the North Carolina mineral (data from improved estimate) and 70 at. % in the Madras mineral.

The distribution of the lanthanons differs markedly from that described for the Ti-rich niobate-tantalates as it is evident that the four samarskites show fairly strong selective assemblage tendencies; Gd, Tb, and Dy are preferentially concentrated and together constitute 72, 70, 54 and 68 at. % of the lanthanons in minerals nos. 11–14 respectively (table II). Only five of minerals nos. 1–10 showed Gd + Tb + Dy exceeding 40 at. % of the lanthanons and the highest percentage was 46 (for no. 9). The data in table II also indicate that lanthanons heavier than Dy are consistently rarer in the lanthanons from the samarskites than in those from the other minerals except for Tm and Lu in the Nouseet, South-west Africa, mineral. The percentages of La, Ce, Pr, Nd, and Sm are comparable.

The samarskite from Mitchell Co., North Carolina, shows a remarkably high at. % of Gd in the lanthanons which almost equals that of all the other lanthanons together (fig. 5 shows the lanthanon distribution); Gd is even more than half as abundant as Yt (25 at. % to 40 at. %). Gd in the mineral is only 4·36 wt. % Gd_2O_3—a figure below that found in a number of gadolinites. Gadolinites, however, are, with respect to the rare earths, minerals of the complete-assemblage type and Gd can be exceeded by other (*Z*-even) lanthanons; moreover, the atomic ratio Yt/Gd is usually greater than 4. To date the samarskite from Mitchell



5. Lanthanon distribution in Ti-poor niobate-tantalate (= samarskite), ell County, North Carolina. B.M. 68781. Points representing the *Z*-even lanthanons are joined.

North Carolina, is unique in its high concentration of Gd relative other rare earths and as a source for either Gd or Tb would be most ble.

The remaining three samarskites have comparable lanthanon di tributions but with Gd about 25 at. % in the lanthanons and Dy ne or slightly higher; Tb ranges from only 6·5 to 7·5 at. % in all fo

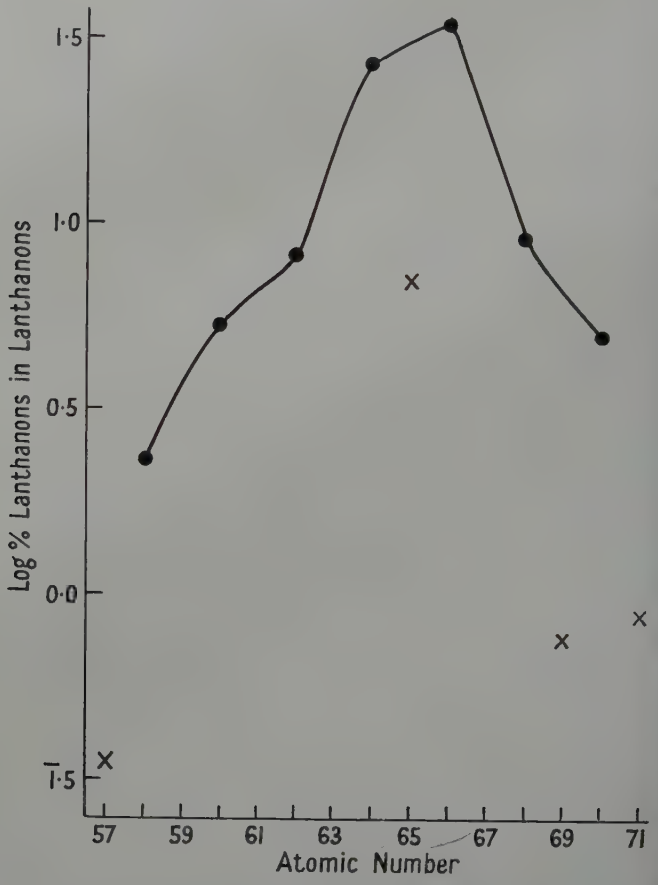


FIG. 6. Lanthanon distribution in Ti-poor niobate-tantalate, Madras, India. Points representing the *Z*-even lanthanons are joined.

samarskites, recalling its limited variation compared with the oth lanthanons (2 to 4·4 at. %) in the Ti-rich niobate–tantalates.

The samarskite from Madras shows the least selective rare ear assemblage of the four samarskites (fig. 6), but its lanthanon distributi is distinct enough from those for minerals nos. 1–10. It is tempting suggest that this selective concentration of Gd, Tb, and Dy and relati

ction of Er, Tm, Yb, and Lu is a property of samarskites in general; t is, that they concentrate Gd, &c., and reject Er, &c., from the ironment in which they crystallize. Contemporaneous crystalliza- of samarskite with davidite could explain the peculiar lanthanon ribution in the latter mineral.

However, more than four analyses are needed before generalizations be more than guessed at; a very Yt-rich (say greater than 90 % of the rare earths) and lanthanon-poor samarskite may well show ore dispersed lanthanon distribution, as those lanthanons differing in and other crystallization properties from Yt would produce cor- ondingly less size strain in the lattice than when total lanthanons e more nearly equal in abundance to Yt.

n conclusion, it is relevant to refer to current research at Imperial ege on rare earths in members of the fergusonite–formanite series, ch to date indicates that lanthanon distribution in fergusonites $_2$,Ln)$_2$O$_3$ > 30 wt. %) is more akin to that in the Ti-rich niobate- alates (nos. 1–10) than to that in samarskites (nos. 11–14). (Wylie, 4, has reported data on an Australian formanite.) In particular a usonite from North Carolina, U.S.A., ((Yt,Ln)$_2$O$_3$ 43·30 wt. %) ved Dy and Er approximately equal and both exceeding Gd; Yt unted for 67 at. % of the rare earths.

knowledgements. I wish to thank Miss Rosemary Hall for carrying out the ical analyses and J. E. T. Horne of H. M. Geological Survey (Atomic Division) nany helpful discussions; Sir William Ogg, Director of Rothamsted Experi- al Station, Harpenden, has again kindly allowed me to use the Mannkopff rograph there; mineral samples have been most generously supplied by the ogisk Museum, Oslo, the British Museum (Natural History), and H. M. Geo- al Survey and Museum.

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rther consideration of the crystal structure of kaolinite.

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[Taken as read 23 January 1958.]

ummary. The layer structure of the kaolin mineral dickite has been shown to er significantly from the idealized arrangement of regular tetrahedra and octa-ra. Since the structure of kaolinite can be studied only by X-ray powder hods, it cannot be refined beyond the idealized arrangement. By applying the ite type of layer structure to the case of kaolinite, an explanation for the inic symmetry of kaolinite has been obtained, and also an improved agreement n the observed X-ray intensities.

A DETAILED structure analysis of kaolinite was first attempted by Gruner (1932), based on X-ray powder diagrams of the mineral. concluded that the unit cell was monoclinic, with probable space up C_s^4–Cc, and contained two structural layers related by a c-glide ne. However, further examination of powder diagrams of various linites by Brindley and Robinson (1946) revealed that there were ections that could not be explained in terms of this monoclinic cell, they put forward a triclinic cell containing only one structural layer. far, it has been proved that this triclinic cell fits very well with the erved features of the X-ray powder patterns of the mineral as regards h spacings and intensities of reflections.

The structure of the composite layer of Si–O and Al–OH was assumed have the geometrically idealized arrangement first proposed by Paul-(1930). At that time, there were no means to establish a refined ucture for the layer itself in the absence of suitable single crystals. In ns of this idealized layer, it is impossible to account in the actual ucture of kaolinite for the α-angle and for a small departure of the ngle from an idealized displacement of $-a/3$.

Recently, Newnham and Brindley (1956, 1957) have made a detailed lysis of the mineral dickite, a polymorphic form of kaolinite, and e shown that the actual layer structure is distorted to some extent n the idealized arrangement. This has opened the way to an inter-tation of the triclinic nature of kaolinite, since there are good reasons thinking that the structures of the layers in dickite and kaolinite

are very nearly the same. The unit-cell parameters (see table I), X-r basal reflections, chemical compositions, infra-red spectrograms, a other data for the two minerals are almost identical.

TABLE I. Comparisons of lattice parameters of kaolinite and dickite.

	Kaolinite Brindley and Robinson (1946).	*Kaolinite* Newnham (1956).	*Dickite* Newnham and Brindley (1956, 1957).
a	5·15 Å.	5·139±0·014 Å.	5·149±0·003
b	8·95	8·932±0·016	8·949±0·005
c	7·39	7·371±0·019	14·419±0·007
α	91·8°	91·6°±0·2°	
β	104·5–105°	104·8°±0·2°	96·80°±0·07°
γ	90°	89·9°±0·1°	
	$d(001) = c(1-\cos^2\alpha-\cos^2\beta)^{\frac{1}{2}} = 7{\cdot}15$ Å.		$d(001)/2 = c(\sin\beta)2 =$ $= 7{\cdot}16$ Å.

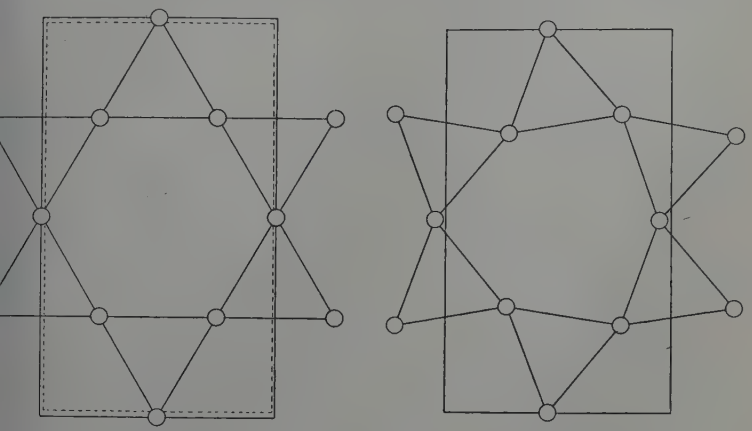
The present paper describes further consideration of the triclin structure of kaolinite based on the layer structure of dickite.

A discussion of the kaolinite structure in relation to the general features the dickite layer.

The outstanding features of the structure of the dickite layer lie in t rotations of both tetrahedra and octahedra of the composite hexagon network (Newnham and Brindley, 1956, figs. 3*a*, 3*b*). In an idealiz layer of Si–O tetrahedra, if the Si–O and O–O distances are assumed be 1·62 Å. and 2·65 Å. respectively, the regular tetrahedral network h a 5·30 and b 9·17 Å. which do not fit with the unit cell dimensions of t kaolin minerals as shown in fig. 1*a*. To fit these dimensions with tho of the kaolin minerals, it is necessary to rotate each tetrahedron paral to the layer in such a manner that the neighbouring tetrahedra rotate opposite directions, as shown in fig. 1*b*. Such rotations have been fou in the actual structure of the dickite layer. The angle of rotation is abo 8° in the dickite layer, which agrees fairly well with an idealized rotati of 10° to produce the actual cell dimensions of the kaolin minerals.

So far, a rotation of octahedra in the network of Al–OH has be found in the structure of gibbsite (Megaw, 1934). The polarization of t hydroxyl ions is affected by the high polarizing power of aluminium io to such an extent as to form hydroxyl bondings between neighbouri hydroxyl ions, resulting in a distortion of the OH-layer from an idealiz arrangement. In the dickite structure this sort of distortion has aga been found together with a slight expansion of the layer dimensions fit with those of the Si–O tetrahedral layer. In an actual layer of dicki

e rotation of triangles composed of three hydroxyl ions in the outer-ost layer is about 3°, while the rotation of these ions in the middle layer 8° in the opposite direction. These rotations result in a shortening of ared edges of octahedra in accordance with Pauling's rules. Further-ore, as compared with the idealized structural layer, the net result of



(*a*) (*b*)

. 1. *a*. Projection on (001) of an idealized silica layer. Dotted lines show the t-cell dimensions of kaolinite. *b*. Projection on (001) of a silica layer with the rahedra rotated to fit the unit-cell dimensions of kaolinite. (Only the oxygen atoms are shown.)

ese rotations is a change of the mutual positions of the outermost ygen and hydroxyl sheets of atoms with regard to the *a*- and *b*-ections.

It can be supposed that the departure of the actual layer from the ometrically idealized arrangement arises from the interactions between s in sde the structural layer, while the stacking of successive layers l be defined mainly by interactions of oxygen ions of one layer with hydroxyl ions of the adjacent layer which form hydroxyl bondings h each other. If we assume the hydroxyl bonds to be equally dis-uted, then the distortion of the kaolin layer modifies the displace-nt of one structural layer with respect to the adjacent layer along h *a*- and *b*-directions. These displacements are shown in figs. 2*a* 2*b*.

ig. 2*a* shows the relation between two adjacent kaolinite layers ing the idealized structure displaced so that the hydroxyl bonds are equal. The layer displacement is $-a/3$ and the β angle is 103·5°.

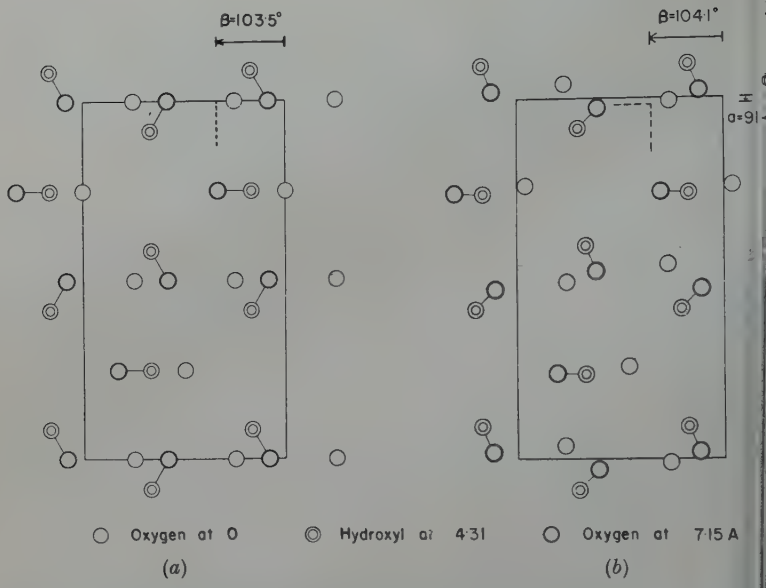


FIG. 2. *a*. Projection on (001) of the idealized kaolinite structure. Dotted lines show the displacement of the adjacent layer required to equalize the OH–O bonds which are shown by short full lines between adjacent oxygen and hydroxyl ions. *b*. Projection on (001) of the kaolinite structure based on the dickite-type layer.

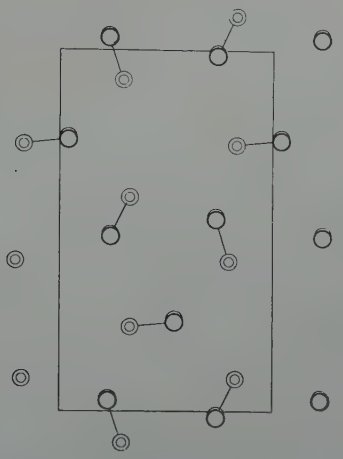


FIG. 3. Projection on (001) of a structure containing dickite-type layers stacked orthogonally.

the other hand, in fig. 2*b* in which a dickite-type layer is used, the ual distribution of OH–O bonds results in the α- and β-angles of olinite being 91·4° and 104·1° respectively which are compatible with e observed values, 91·6° for α and 104·8° for β. In the structure of kite itself, the displacements along *b* are cancelled out throughout the ole crystal, owing to the overall monoclinic symmetry resulting from e two layers in the cell.

It is of interest also to consider the bearing of the dickite-type layer ucture on the various layer sequences that have been shown to be ssible for the idealized layer (Brindley, 1951). On the basis of equal H–O bonds between the layers, many stacking sequences appear to be ually probable. The simplest arrangement is an orthogonal one that lds exactly the same distribution of OH–O bonds as is depicted in

BLE II. Comparisons between calculated and observed X-ray intensities for kaolinite.

	Calculated						Observed	
Indices	I.		II.		III.		IV.	V.
(020)	69		75		104		13	m−
(1$\bar{1}$0)	200		205		224		27	m
(110)	20		7		2		—	—
(11$\bar{1}$)	210		219		213		23	m
(1$\bar{1}\bar{1}$)	*100*		*100*		*100*		*10*	w+
(02$\bar{1}$)	163		164		108		9	w−m
(021)	85		50		51		4	w
(1$\bar{3}$0)	27		32		42			
(20$\bar{1}$)	26	*100*	34	*100*	38	*100*	*10*	ms
(130)	47		34		20			
(13$\bar{1}$)	54	95	61	74	59	89	6	w−m
(1$\bar{1}$2)	41		13		30			
(13$\bar{1}$)	67		73		78			
(200)	64	146	84	169	69	161	13	s
(112)	15		12		14			
(20$\bar{2}$)	187	380	102	218	90	175	20	vs
(13$\bar{1}$)	193		116		85			
(11$\bar{3}$)	28	218	19	140	10	108	11	s
(131)	190		121		98			

ls. I, II, and III give calculated values of $F^2\ \Phi(\theta)$, where $\Phi(\theta)$ is the polarization Lorentz factor. For reflections with $k = 3n$, the line (1$\bar{3}$0), (20$\bar{1}$), (130) is taken standard; with $k \neq 3n$, (11$\bar{1}$) is taken as a standard.

l. I is for Brindley and Robinson's structure based on the idealized layer, II for the structure based on the dickite-type layer, and col. III for a trial ture based on a somewhat more distorted layer than that in dickite.

l. IV gives averaged observed X-ray diffractometer data for well-crystallized nites, and col. V gives the qualitative intensities listed by Brindley and nson (1946).

fig. 2*a*. When the dickite-type layer is used the distribution illustrate in fig. 3 is obtained, and it is evident by comparing figs. 2*b* and 3 tha for a given value of $d(001)$, the arrangement 2*b* leads to the shorter OH– bonds. This kind of argument brings one to the point where it is easy t see that different stacking arrangements may lead to secondary varia tions in the detailed structure of the layers and in the bonding betwee the layers. In the absence of suitable data for a precision determinatio of the structure of kaolinite, this line of argument cannot be pursue further for the time being.

In table II, calculations of intensities for kaolinite based on th dickite-type layer are shown along with those for the Brindley–Robinso structure. The agreement with the observed values is somewhat im proved as compared with the earlier calculations. A little more improve ment may be possible by distorting the layer further (see table II, co III), but in view of the small number of unique reflections availab from X-ray powder diagrams it seems meaningless to attempt furth refinement.

Acknowledgements. This work was made possible by a grant-in-aid from th Gulf Research and Development Company, Harmarville, Pennsylvania, U.S.A.

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Third supplementary list of British minerals.

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[Read 7 November 1957.]

MANUAL of the mineralogy of Great Britain and Ireland' by Robert Philips Greg[1] and William G. Lettsom[2] (London, 1858, +483 pp., 400 figs.) still remains the standard work of reference for itish minerals, but is now much out of date and also out of print. was able to obtain in 1895 a second-hand copy, and, with the too ıbitious idea of producing a revised edition, I started collecting ıterial, which soon became overwhelming. So in the meantime I gave 1898 a supplementary list[3] of the names of British species and varieties ıt had been recorded since 1858. After a period of 33 years a second)plementary list[4] was given in 1931. Rather than wait for another 33 ırs, the present third list is offered on the occasion of the centenary Greg and Lettsom'. In addition to these there are many references British minerals in the International Catalogue of Scientific Litera- e (1901–14), and in the indexes to the Mineralogical Magazine and,)e 1920, to Mineralogical Abstracts.

n these three supplementary lists about 250 species have been added he 241 given by Greg and Lettsom in 1858, making a total of 490, as

Robert Philips Greg (1826–1906), biography with portrait in Min. Mag., 1907, 14, pp. 268–271. His mineral collection, which included that of Thomas Allan 7–1833), was purchased for the British Museum in 1860, together with the 3-vol. n-Greg manuscript catalogue.

William Garrow Lettsom (1805-87) was in the diplomatic service in many tries. Biographical note in Monthly Notices Roy. Astron. Soc., 1888, vol. 48, 165–166. His mineral collection passed to William Nevill of Godalming and to Charles Hampton Turner of Godstone, Surrey. These all came into the ction of Henry Ludlam, which in 1880 he bequeathed to the Museum of Practi- eology (now Geological Survey Museum). The collection of Lettsom's grand- er, John Coakley Lettsom (1744-1815), was the foundation in 1793 of the ralogical Museum of Harvard University.

. J. Spencer, Supplementary list of British minerals. 68th Report Brit. Assoc. Sci., 1899 (for 1898), pp. 875–877; reprinted in Geol. Mag., 1899, decade 4, vol. . 75–76; Zeits. Kryst. Min., 1900, vol. 32, pp. 287–289.

econd supplementary list of British minerals. Report Brit. Assoc. Adv. Sci., (for 1931), centenary vol., London, p. 378 [M.A. 5–85].

compared with 824 numbered species in Dana's sixth edition in 189 and now, due to X-ray methods, say 1 300.

Adamite Zn_2AsO_4OH Orthorhombi

From several localities in Cumberland and Cornwall. A. W. G. King bury, Min. Soc. Notice, 1958, no. 100.

Afwillite $Ca_3(SiO_3OH)_2.2H_2O$ Monoclin

In chalk–dolerite contact-zone, Scawt Hill, Larne, Co. Antrim. C. H Tilley, Geol. Mag., 1930, vol. 69, p. 168 [M.A. **4**–330]; Min. Mag., 193 vol. 22, p. 444; H. D. Megaw, Acta Cryst., 1952, vol. 5, p. 477 [M.A **12**–98].

Aikinite $CuPbBiS_3$ Orthorhomb

In greisen, Grainsgill, Carrock Fell, Cumberland. A. W. G. King bury and J. Hartley, Min. Mag., 1956, vol. 31, p. 297.

Alabandine MnS Cub

With rhodochrosite in a mineral veinlet, Tintagel, Cornwall. C. H Tilley, 1944 (private communication).

Alleghanyite $2Mn_2SiO_4.Mn(OH,F)_2$ Monoclin

In metamorphosed manganese ore, Benallt mine, Rhiw, Carnarvo shire. W. C. Smith, F. A. Bannister, and M. H. Hey, Min. Mag., 194 vol. 27, p. 40.

Antlerite $CuSO_4.2Cu(OH)_2$ Orthorhomb

Lake District, Cumberland. A. W. G. Kingsbury and J. Hartle Min. Soc. Notice, 1951, no. 72 [Amer. Min. **36**–637].

Old Gunnislake mine, Calstock, Cornwall. A. W. G. Kingsbury, Tran Roy. Geol. Soc. Cornwall, 1954, vol. 18, pt. 4 (for 1952), p. 338 [M.A **12**–362].

Aphthitalite $(K,Na)_2SO_4$ Rhombohedr

In salt deposits, Whitby, Yorkshire. F. H. Stewart, Proc. York Geol. Soc., 1954, vol. 29, p. 228 [M.A. **12**–499].

Arseniosiderite $Ca_3Fe_4(AsO_4)_4(OH)_6.3H_2O$

Cumberland (four localities), Cornwall, and Devonshire. A. W. Kingsbury and J. Hartley, Min. Mag., 1957, vol. 31, p. 499.

Banalsite $Na_2BaAl_4Si_4O_{16}$ Orthorhombic

In metamorphosed manganese ore, Benallt mine, Rhiw, Carnarvonshire. W. C. Smith, F. A. Bannister, and M. H. Hey, Min. Mag. 1944, vol. 27, p. 33; W. C. Smith, ibid., 1945, vol. 27, p. 63.

Barbertonite $Mg_6Cr_2(OH)_{16}CO_3.4H_2O$ Hexagonal

The mineral described as stichtite (q.v.) may represent the hexagonal rather than the rhombohedral modification. C. Frondel, Amer. Min., 1941, vol. 26, p. 311 [M.A. **8**–100].

Basaluminite $2Al_2O_3.SO_3.10H_2O$

White, compact, filling fissures in ironstone, Irchester, Northamptonshire, and in chalk, Clifton Hill, Brighton, Sussex. F. A. Bannister and S. E. Hollingworth, Nature, London, 1948, vol. 162, p. 565 [M.A. 10–452]; Hollingworth and Bannister, Min. Mag., 1950, vol. 29, p. 1. (*See* Hydrobasaluminite.)

Bavenite $Ca_4(Be,Al)_4Si_9(O,OH)_{28}$ Monoclinic

In aplite, Meldon, Devonshire. W. R. Dearman and G. F. Claringbull, 1956 (private communication).

Beaverite $Pb(Cu,Fe,Al)_3(SO_4)_2(OH)_6$ Rhombohedral

From Cumberland and Cornwall. A. W. G. Kingsbury, Min. Mag., 1953, vol. 30, p. xliv; Trans. Roy. Geol. Soc. Cornwall, 1954, vol. 18, pt. 4 (for 1952), p. 394 [M.A. **12**–362]; Min. Mag., 1957, vol. 31, p. 700.

Bementite $2MnSiO_3.H_2O$ (?)

In manganese ore, Treburland mine, Altarnun, Cornwall. A. Russell, Min. Mag., 1946, vol. 27, p. 225.

Beudantite $PbFe_3AsO_4SO_4(OH)_6$ Rhombohedral

Cumberland. A. W. G. Kingsbury, Min. Soc. Notice, 1954, no. 84.

Penberthy Croft mine, St. Hilary, Cornwall. A. W. G. Kingsbury, Trans. Roy. Geol. Soc. Cornwall, 1954, vol. 18, pt. 4 (for 1952), p. 392 [M.A. **12**–362]. (*See* Corkite.)

Beberite $CoSO_4.7H_2O$ Monoclinic

Penberthy Croft mine and Wheal Alfred, Cornwall. J. Robson, Trans. Roy. Geol. Soc. Cornwall, 1949, vol. 17, pt. 8 (for 1948), p. 472 [M.A. **11**–273]. Previously mistaken for erythrite.

Boehmite $AlO(OH)$ Orthorhombic

In bauxitic clay, Kilwinning, Ayrshire. J. de Lapparent, Compt. Rend. Acad. Sci. Paris, 1934, vol. 199, p. 1629; Bull. Soc. Franç. Min., 1935, vol. 58, p. 246; Summ. Rep. Progr. Geol. Survey Great Britain for 1934, pt. 2, p. 1 [M.A. **6**–135, 373].

In bauxitic clay baked by lava flows or basalt dikes, Co. Antrim. V. A. Eyles, Mem. Geol. Surv. Northern Ireland, 1952 [M.A. **12**–159].

Boracite $Mg_6Cl_2B_{14}O_{26}$ Pseudo-cubic

In salt deposits, Aislaby, Eskdale, Yorkshire. E. M. Guppy, Min. Mag., 1944, vol. 27, p. 51; F. H. Stewart, ibid., 1951, vol. 29, p. 449.

Boulangerite $Pb_5Sb_4S_{11}$ Monoclinic

In greisen, Grainsgill, Carrock Fell, Cumberland. A. W. G. Kingsbury and J. Hartley, Min. Mag., 1956, vol. 31, p. 297.

Endellion, Cornwall. Ibid., p. 299.

Brammallite Sodium-illite

In coal-measure shales, Llandebie, Carmarthenshire. F. A. Bannister, Min. Mag., 1943, vol. 26, p. 304. (*See* Illite.)

Bravoite $(Fe,Ni)S_2$ Cubic

Mill Close mine, Derbyshire. F. A. Bannister, Min. Mag., 1940, vol. 25, p. 609.

Bredigite α'-Ca_2SiO_4 Orthorhombic

Chalk–dolerite contact-zone, Scawt Hill, Larne, Co. Antrim. C. E. Tilley and H. C. G. Vincent, Min. Mag., 1948, vol. 28, p. 255; A. M. B. Douglas, ibid., 1950, vol. 29, p. 875.

Chalk–dolerite contact-zone, Ballycraigy, Larne, Co. Antrim. J. D. C. McConnell, Min. Mag., 1954, vol. 30, pp. 302, 672.

Limestone–gabbro contact-zone, Camas Mòr, Isle of Muck, Inverness-shire. C. E. Tilley, Min. Mag., 1947, vol. 28, p. 257 (footnote).

Gabbro intrusion in limestone, Camphouse, Ardnamurchan, Argyllshire. S. O. Agrell, Min. Soc. Notice, 1950, no. 69 [Amer. Min., **38**–1080].

Bustamite $(Mn,Ca,Fe)SiO_3$ Triclinic

Manganese ore, Treburland mine, Altarnun, Cornwall. C. E. Tilley, Min. Mag., 1946, vol. 27, p. 236; A. Russell, ibid., p. 224.

ancrinite, sulphatic $(Na,K,Ca)_{6-8}(Si,Al)_{12}O_{24}(SO_4,CO_3)_{1-2}.nH_2O$ Hexagonal

In borolanite, Loch Borolan, Assynt, Sutherlandshire. F. H. Stewart, Min. Mag., 1941, vol. 26, p. 1.

Carnallite $KMgCl_3.6H_2O$ Orthorhombic

In salt deposits, Aislaby, Eskdale, Yorkshire. F. H. Stewart, Min. Mag., 1951, vol. 29, pp. 449, 557; 1956, vol. 31, p. 127; G. Armstrong *et al.*, ibid., 1951, vol. 29, p. 667; L. R. Raymond, Quart. Journ. Geol. Soc. London, 1953, vol. 108 (for 1952), p. 289 [M.A. **12**–283].

Cerpholite $MnAl_2Si_2O_6(OH)_4$ Monoclinic

Grainsgill, Carrock Fell, Cumberland, and three localities in Cornwall. W. G. Kingsbury and J. Hartley, Min. Mag., 1957, vol. 31, p. 502.

Cheruleite $CuO.2Al_2O_3.As_2O_5.8H_2O$

Sky-blue, botryoidal, on liroconite, Wheal Gorland, Gwennap, Cornwall. A. Russell and G. F. Claringbull, Min. Soc. Notice, 1955, no. 90 [M.A. **13**–30].

Chinohumite $Mg_9Si_4O_{16}(F,OH)_2$ Monoclinic

With humite and chondrodite in dolomite–granite contact-zone, Loadford, Skye, Inverness-shire. C. E. Tilley, Geol. Mag., 1948, vol. 85, p. 215 [M.A. **10**–355]; Min. Mag., 1951, vol. 29, p. 636.

Collophane $Ca_3(PO_4)_2.H_2O$

In manganese ore, Benallt mine, Carnarvonshire. A. W. Woodland, Proc. Geol. Assoc., 1939, vol. 50, p. 213 [M.A. **8**–250].

In magnesian limestone, Castle Eden, Co. Durham. K. C. Dunham, F. Claringbull, and F. A. Bannister. Min. Mag., 1948, vol. 28, p. 338.

In Thames gravel, London. S. E. Ellis and G. F. Claringbull, Min. Mag., 1951, vol. 29, p. 615.

In chalk, Princes Risborough, Buckinghamshire. G. Brown and C. D. Pilcher, Min. Mag., 1956, vol. 31, p. 339.

With bones, Tornewton Cave, Devonshire. V. B. Proudfoot, Min. Mag., 1956, vol. 31, p. 344.

Columbite $(Fe,Mn)(Nb,Ta)_2O_6$ Orthorhombic

In aplite, Meldon, Devonshire. O. von Knorring, Min. Mag., 1951, vol. 29, p. 799.

Conichalcite $CuCaAsO_4(OH)$ Orthorhomb

Cornwall. A. W. G. Kingsbury and J. Hartley, Min. Soc. Notice, 195 no. 74 [Amer. Min., **36**–784].

Bedford United mines, Tavistock, Devonshire. A. W. G. Kingsbur Trans. Roy. Geol. Soc. Cornwall, 1954, vol. 18, pt. 4 (for 1952), p. 39

Cookeite $LiAl_4Si_3AlO_{10}(OH)_2$ Monoclin

In quartz–sulphide ore with gold, Ogofau, Carmarthenshire. Brammall, J. G. C. Leech, and F. A. Bannister, Min. Mag., 1937, vo 24, p. 507.

Corkite $PbFe_3(PO_4)(SO_4)(OH)_6$ Rhombohedr

In iron ore, Glendore mine, Co. Cork. Previously referred to beudan ite (q.v.) by Greg and Lettsom from which it differs in having (PO_4) place of (AsO_4).

Cornubite $Cu_5(AsO_4)_2(OH)_4$

With cornwallite on quartz, Wheal Carpenter, Gwinear, Cornwa G. F. Claringbull, M. H. Hey, and A. Russell, Min. Soc. Notice, 1957, n 99; Min. Abstr., 1958, vol. 13, p. 558.

Cosalite $CuPb_7Bi_8S_{22}$ Orthorhomb

Fibrous tufts in greisen, Grainsgill, Carrock Fell, Cumberlan A. W. G. Kingsbury and J. Hartley, Min. Mag., 1956, vol. 31, p. 297.

Crossite $(Na,K,Ca)_{2\cdot7}(Mg,Fe,Al,Ti)_5(Si,Al)_8O_{23}(OH)$ Monoclin

A blue soda-amphibole previously referred to glaucophane, in 'glauc phane'-schist, Llanfairpwllgwyngyll, Anglesey, Carnarvonshire. Holgate, Min. Mag., 1951, vol. 29, p. 792.

Cubanite $CuFe_2S_3$ Orthorhomb

In veins of tin ore, East Pool mine, Redruth, Cornwall. J. C. Brow Neues Jahrb. Min. Abt. A., 1934, Beil.-Bd. 68, p. 321 [M.A. **6**–228].

Cuspidine $Ca_4Si_2O_7F_2$ Monoclin

'Custerite' in metamorphosed limestone, Carlingford, Co. Lout G. D. Osborne, Geol. Mag., 1933, vol. 69, pp. 61, 219 [M.A. **5**–134]; S. H Nockolds, Min. Mag., 1947, vol. 28, p. 157.

In dolomite–granite contact-zone, Broadford, Skye, Inverness-shir C. E. Tilley, Min. Mag., 1947, vol. 28, p. 90; 1951, vol. 29, p. 634.

In limestone–gabbro contact-zone, Camas Mòr, Isle of Muck, Inverness-shire. C. E. Tilley, Bull. Comm. Géol. Finlande, 1947, no. 140, p. 100 [M.A. **10**–258].

Cyanotrichite $Cu_4Al_2SO_4(OH)_{12}.2H_2O$ Orthorhombic

[Synonym, lettsomite (J. Percy, 1850) from Moldava, Banat.]

Leadhills, Lanarkshire. C. Frondel (1949) in Dana's Min., 7th edn., 1951, p. 579.

Wheal Gorland, St. Day, Cornwall. A. W. G. Kingsbury and J. Hartley, Min. Soc. Notice, 1951, no. 74 [Amer. Min., **36**–784]; A. W. G. Kingsbury, Trans. Roy. Geol. Soc. Cornwall, 1954, vol. 18, pt. 4 (for 1952), p. 389 [M.A. **12**–362].

Cymrite $BaAlSi_3O_8(OH)$ Hexagonal

In metamorphosed manganese ore, Benallt mine, Rhiw, Carnarvonshire. W. C. Smith, F. A. Bannister, and M. H. Hey, Min. Mag., 1949, vol. 28, p. 676.

Dickite $Al_2Si_2O_5(OH)_2$ Monoclinic

Amlwch, Anglesey, Carnarvonshire. C. S. Ross and P. F. Kerr, Amer. Min., 1930, vol. 15, p. 34 [M.A. **4**–247]. Described as kaolinite by A. B. Dick and H. A. Miers, Min. Mag., 1888, vol. 8, pp. 15, 24.

In sandstones, northern England and North Wales. F. Smithson and S. Brown, Min. Mag., 1957, vol. 31, p. 381.

Duftite $PbCuAsO_4(OH)$ Orthorhombic

Brandy Gill, Carrock Fell, Cumberland. G. F. Claringbull, Min. Mag., 1951, vol. 29, p. 609; W. F. Davidson and N. Thomson, 1951 [M.A. 12–384].

Erythrosiderite $K_2FeCl_5.H_2O$ Orthorhombic

In salt deposits, alteration product of rinneïte, Aislaby, Eskdale, Yorkshire. F. H. Stewart, Min. Mag., 1951, vol. 29, p. 563.

Ettringite $Ca_6Al_2(SO_4)_3(OH)_{12}.26H_2O$ Hexagonal

In chalk–dolerite contact-zone, Scawt Hill, Larne, Co. Antrim. C. E. Tilley and H. F. Harwood, Min. Mag., 1931, vol. 22, p. 444; F. A. Bannister, ibid., 1936, vol. 24, p. 324.

Euclase $BeAlSiO_4OH$ Monoclinic

Cligga Head, Perranporth, Cornwall. A. W. G. Kingsbury, Min. Mag., 1958, vol. 31, p. 815.

Eudialyte $Na_4(Ca,Fe)_2ZrSiO_{17}(OH,Cl)_2$ Rhombohedral

In quartz-syenite veins in limestone, Barnavave, Carlingford, Co. Louth. S. R. Nockolds, Min. Mag., 1950, vol. 29, p. 27.

Eudnophite $NaAlSi_2O_6.H_2O$ Pseudo-cubic

Variety of analcime, in borolanite, Loch Borolan, Assynt, Sutherlandshire. F. H. Stewart, Min. Mag., 1941, vol. 26, p. 1. (*See* Leucite.)

Ferrobrucite $(Mg,Fe)(OH)_2$ Rhombohedral

Variety of brucite, in limestone–gabbro contact-zone, Isle of Muck, Inverness-shire. C. E. Tilley, Bull. Comm. Géol. Finlande, 1947, no. 140, p. 102 [M.A. **10**–235].

Fluoborite $Mg_3BO_3(F,OH)_3$ Hexagonal

In dolomite–granite contact-zone, Broadford, Skye, Inverness-shire, C. E. Tilley, Geol. Mag., 1948, vol. 85, 215 [M.A. **10**–355]; Min. Mag., 1951, vol. 29, p. 637.

Ganophyllite $Mn_7Al_2Si_8O_{26}.6H_2O$ Monoclinic

In manganese ore, Benallt and Nant mines, Rhiw, Carnarvonshire. W. C. Smith, F. A. Bannister, and M. H. Hey, Min. Mag., 1946, vol. 27, p. 218; ibid., 1948, vol. 28, p. 343.

Gedrite $(Mg,Fe'',Al)_7(Si,Al)_8O_{22}(OH)_2$ Orthorhombic

In schists and granulite, Strathy, Sutherlandshire. R. S. Collins, Min. Mag., 1942, vol. 26, p. 254.

Gedritite rock between serpentinite and kyanite–hydrobiotite rock, Glen Urquhart, Inverness-shire. G. H. Francis, Min. Mag., 1955, vol. 30, p. 709.

Gehlenite $Ca_2Al_2SiO_7$ Tetragonal

In chalk–dolerite contact-zone, Scawt Hill, Larne, Co. Antrim. C. E. Tilley, Min. Mag., 1929, vol. 22, p. 83.

In limestone–gabbro contact-zone, Camas Mòr, Isle of Muck, Inverness-shire. C. E. Tilley, Bull. Comm. Géol. Finlande, 1947, no. 140, p. 99 [M.A. **10**–285].

Gabbro intrusion in limestone, Camphouse, Ardnamurchan, Argyllshire. S. O. Agrell, Min. Soc. Notice, 1950, no. 69 [Amer. Min. **38**–1080].

Gonnardite $(Ca,Na)_{6-8}(Si,Al)_{20}O_{40}.12H_2O$ Orthorhombic (?)

With saponite in vesicular basalt, Allt Ribhein, Fiskavaig Bay, Skye, Inverness-shire. R. C. Mackenzie, Min. Mag., 1957, vol. 31, p. 672.

Greenalite $3FeO.4SiO_2.2H_2O$ Isotropic

In greenalite-chert, Glenluce, Wigtownshire. W. Q. Kennedy, Min. Mag., 1936, vol. 24, p. 433.

Grovesite $(Mn,Mg,Al)_2(Si,Al)_2(O,OH)_9$

In metamorphosed manganese ore, Benallt mine, Rhiw, Carnarvonshire. F. A. Bannister, M. H. Hey, and W. C. Smith, Min. Mag., 1955, vol. 30, p. 645. Closely allied to pennantite (q.v.).

Grunerite $Fe_7Si_8O_{22}(OH)_2$ Monoclinic

In greenstone-hornfels with almandine, Botallack, Cornwall. C. E. Tilley, Min. Mag., 1935, vol. 24, p. 191; A. R. Alderman, ibid., p. 42.

In eulysite, Loch Duich, Ross-shire. C. E. Tilley, Min. Mag., 1936, vol. 24, p. 333.

In pyroxmangite–grunerite–garnet schist, Glen Beag, Glenelg, Inverness-shire. C. E. Tilley, Amer. Min., 1937, vol. 22, p. 720 [M.A. **6**–528].

Harkerite Ca–Mg carbonate borosilicate Cubic

In dolomite–granite contact-zone, Broadford, Skye, Inverness-shire. C. E. Tilley, Min. Mag., 1949, vol. 28, proc. p. lxxi (for 1948); Geol. Mag., 1948, vol. 85, p. 215 [M.A. **10**–355]; Min. Mag., 1951, vol. 29, p. 640.

Hedenbergite $(Ca,Fe)SiO_3$ Monoclinic

In metamorphosed limestone, Tregullen, Bodmin, Cornwall. G. Barrow and H. H. Thomas, Min. Mag., 1908, vol. 15, p. 117.

In eulysite, Loch Duich, Ross-shire. C. E. Tilley, Min. Mag., 1936, vol. 24, p. 334.

Dolomite–granite contact-zone, Broadford, Skye, Inverness-shire. C. E. Tilley, Geol. Mag., 1948, vol. 85, p. 214 [M.A. **10**–354]; Amer. Min., 1948, vol. 33, p. 736 [M.A. **11**–16]; Min. Mag., 1951, vol. 24, p. 635.

Herderite $CaBePO_4F$ Orthorhombic

Wheal Cock, St. Just, Cornwall, and Colcerrow quarry, Luxullyan, Cornwall. A. W. G. Kingsbury, Min. Mag., 1958, vol. 31, p. 816.

Hillebrandite $Ca_2SiO_4.H_2O$ Orthorhombic

In metamorphosed limestone, Carlingford, Co. Louth. S. R. Nockolds, Min. Mag., 1947, vol. 28, p. 156.

Hübnerite $MnWO_4$ Monoclinic

Grainsgill, Carrock Fell, Cumberland. A. W. G. Kingsbury, Min. Soc. Notice, 1958, no. 100.

Hydrobasaluminite $2Al_2O_3.SO_3.41H_2O$

Plastic material containing more water than basaluminite (q.v.). F. A. Bannister and S. E. Hollingworth, Nature, London, 1948, vol. 162, 565 [M.A. **10**–452]; Hollingworth and Bannister, Min. Mag., 1950, vol. 29, p. 1.

Hydrobiotite Hydrated biotite grading to vermiculite.

In amphibolite, Malvern, Worcestershire. A. Brammall, J. G. C. Leech, and F. A. Bannister, Min. Mag., 1937, vol. 24, p. 517.

In soil-clays, Aberdeenshire. G. W. Walker, Min. Mag., 1949, vol. 28, p. 699.

Hydrocalumite $4CaO.Al_2O_3.12H_2O$ Monoclinic

In chalk–dolerite contact-zone, Scawt Hill, Larne, Co. Antrim. C. E. Tilley, Min. Mag., 1934, vol. 23, p. 607.

Hydrogrossular $Ca_3Al_2[SiO_4,4(OH)]_3$ Pseudo-cubic

In limestone–gabbro contact-zone, Camas Mòr, Isle of Muck, Inverness-shire. C. E. Tilley, Bull. Comm. Géol. Finlande, 1947, no. 140, p. 99 [M.A. **10**–285].

Hydrogarnet in metamorphosed limestone, Camphouse, Ardnamurchan, Argyllshire. S. O. Agrell, Min. Soc. Notice, 1950, no. 69 [Amer. Min., **38**–1080].

Hydromuscovite Hydrated muscovite grading to illite (q.v.).

In quartz–sulphide ore with gold, Ogofau, Carmarthenshire. A. Brammall, J. G. C. Leech, and F. A. Bannister, Min. Mag., 1937, vol. 24, p. 507.

Illite Finely divided hydrated muscovite. Monoclinic

In soil-clay, Aberdeenshire. G. W. Walker, Min. Mag., 1950, vol. 29, p. 75.

In fireclay, Yorkshire. K. Carr *et al.*, Min. Mag., 1953, vol. 30, p. 139.

In Old Red Sandstone soils, Denholm Hill quarry, Roxburghshire. R. C. Mackenzie, Min. Mag., 1957, vol. 31, p. 681.

See Brammallite 'sodium–illite'.

Iron-wollastonite $(Ca,Fe)SiO_3$ Monoclinic

In chalk–dolerite contact–zone, Scawt Hill, Larne, Co. Antrim. C. E. Tilley, Min. Mag., 1937, vol. 24, p. 572.

In limestone–gabbro contact-zone, Camas Mòr, Isle of Muck, Inverness-shire. Bull. Comm. Géol. Finlande, 1947, no. 140, p. 103 [M.A. **10**–285].

In dolomite–granite contact-zone, Broadford, Skye, Inverness-shire. C. E. Tilley, Amer. Min., 1948, vol. 33, p. 736 [M.A. **11**–16]; Min. Mag., 1951, vol. 29, p. 634.

Ferrowollastonite in metamorphosed limestone, Camphouse, Ardnamurchan, Argyllshire. S. O. Agrell, Min. Soc. Notice, 1950, no. 69 [Amer. Min., **38**–1080].

Isostannite Cu_2FeSnS_4 Cubic

Cligga Head, Perranporth, Cornwall. G. F. Claringbull and M. H. Hey, Min. Soc. Notice, 1955, no. 91; M. H. Hey, Mineral Index (Brit. Mus.), 1955, pp. 34, 468.

Jacobsite $(Mn,Fe,Mg)(Fe,Mn)_2O_4$ Cubic

In manganese ore, Benallt mine, Rhiw, Carnarvonshire. W. C. Smith, F. A. Bannister, and M. H. Hey, Min. Mag., 1944, vol. 27, p. 39.

Jarosite $KFe_3(SO_4)_2(OH)_6$ Rhombohedral

Several localities in Cumberland. A. W. G. Kingsbury and J. Hartley, Min. Mag., 1958, vol. 31, p. 813.

Jordanite $Pb_{14}As_7S_{24}$ (?) Monoclinic

Penberthy Croft mine, St. Hilary, Cornwall. J. Robson, Trans. Roy. Geol. Soc. Cornwall, 1949, vol. 17, pt. 8 (for 1948), p. 457 [M.A. **11**–273].

Kasolite $PbO.UO_3.SiO_2.H_2O$ Monoclinic

With pitchblende, Tyndrum, Perthshire. R. K. Harrison, Min. Soc. Notice, 1956, no. 95; Min. Mag., 1957, vol. 31, proc. p. lxiv.

Kieserite $MgSO_4.H_2O$ Monoclinic

In salt deposits, Sleights, Eskdale, Yorkshire. G. Armstrong *et al.*, Min. Mag., 1951, vol. 29, p. 672; L. R. Raymond, Quart. Journ. Geol. Soc., 1953, vol. 108 (for 1952), p. 284 [M.A. **12**–283].

Knebelite $(Fe,Mn)_2SiO_4$ Orthorhomb

In quartz–knebelite–magnetite schist, Gairloch, Ross-shire. C. Tilley, Geol. Mag., 1938, vol. 75, p. 77 [M.A. **7**–191].

Langbeinite $K_2SO_4.2MgSO_4$ Cub

In salt deposits, Sleights, Eskdale, Yorkshire. G. Armstrong *et a* Min. Mag. 1951, vol. 29, p. 674.

Larnite β-Ca_2SiO_4 Monoclin

In chalk–dolerite contact-zone, Scawt Hill, Larne, Co. Antrim. C. T Tilley, Min. Mag., 1929, vol. 22, p. 77; ibid., 1931, vol. 22, p. 443.

In chalk–dolerite contact-zone, Ballycraigy, Larne, Co. Antrim J. D. C. McConnell, Min. Mag., 1954, vol. 30, pp. 302, 672.

In limestone–gabbro contact-zone, Camas Mòr, Isle of Muck, Inverness shire. C. E. Tilley, Bull. Comm. Géol. Finlande, 1947, no. 140, p. 10 [M.A. **10**–285].

In metamorphosed limestone, Camphouse, Ardnamurchan, Argyll shire. S. O. Agrell, Min. Soc. Notice, 1950, no. 69 [Amer. Min., **38** 1080].

Leucite $KAl(SiO_3)_2$ Pseudo-cubi

No record found for fresh leucite. Usually assumed to be represente by its alteration product analcime.

In analcime–basalt, Rathjordan, Co. Limerick. G. T. Prior, Min Mag., 1910, vol. 15, p. 315.

Leucite–basanite, leucite–kulaite, &c., East Lothian. E. B. Bailey Mem. Geol. Surv. Scotland, 1910, 126; D. Balsillie, Geol. Mag., 1936, vol 73, p. 16 [M.A. **7**–39]; J. A. E. Bennett, Trans. Edinburgh Geol. Soc. 1945, vol. 14, p. 34 [M.A. **9**–272].

Leucite-minette, Permian lavas, Exeter, Devonshire. W. G. Tid marsh, Quart. Journ. Geol. Soc. London, 1932, vol. 88, p. 755.

Pseudo-leucite in borolanite, Loch Borolan, Sutherlandshire. J. J. H Teall, Geol. Mag., 1900, decade 4, vol. 7. p. 388. (*See* Eudnophite.)

Liebigite $Ca_2U(CO_3)_4.10H_2O$ Orthorhombic

Wheal Basset, Redruth, Cornwall. H. T. Evans and C. Frondel Amer. Min., 1950, vol. 35, p. 252 [M.A. **11**–229].

Lindgrenite $Cu_3(MoO_4)_2(OH)_2$ Monoclinic

Brandy Gill, Carrock Fell, Cumberland. A. W. G. Kingsbury and J. Hartley, Min. Mag., 1955, vol. 30, p. 723 [M.A. **12**–505].

Lizardite $H_4Mg_3Si_2O_9$ Monoclinic

A serpentine mineral, Lizard, Cornwall. E. J. W. Whittaker and J. Zussman, Min. Mag., 1956, vol. 31, p. 118; J. Zussman *et al.*, Amer. Min., 1957, vol. 42, p. 135.

Ludwigite $(Mg,Fe'')_2Fe'''BO_5$ Orthorhombic

In dolomite–granite contact-zone, Broadford, Skye, Inverness-shire. C. E. Tilley, Geol. Mag., 1948, vol. 85, p. 215 [M.A. **10**–355]; Min. Mag., 1951, vol. 29, p. 638.

Manganfayalite $(Fe,Mn)_2SiO_4$ Orthorhombic

In eulysite, Loch Duich, Ross-shire. C. E. Tilley, Min. Mag., 1936, vol. 24, p. 332.

Manganhedenbergite $(Ca,Fe,Mn,Mg)SiO_3$ Monoclinic

In manganese ore, Treburland mine, Altarnun, Cornwall. C. E. Tilley, Min. Mag., 1940, vol. 27, p. 237.

Merwinite $Ca_3Mg(SiO_4)_2$ Monoclinic

In chalk–dolerite contact-zone, Scawt Hill, Larne, Co. Antrim. C. E. Tilley, Min. Mag., 1929, vol. 22, p. 24.

In limestone–gabbro contact-zone, Camas Mòr, Isle of Muck, Inverness-shire. C. E. Tilley, Bull. Comm. Géol. Finlande, 1947, no. 140, p. 100 [M.A. **10**–285].

Metagreenalite $3FeO.4SiO_2.2H_2O$ Anisotropic

In greenalite-chert, Glenluce, Wigtownshire. W. Q. Kennedy, Min. Mag., 1936, vol. 24, p. 434.

Monticellite $CaMgSiO_4$ Orthorhombic

In metamorphosed limestone, Carlingford, Co. Louth. G. D. Osborne, Geol. Mag., 1932, vol. 69, pp. 61, 219 [M.A. **5**–134]; S. R. Nockolds, Min. Mag., 1947, vol. 28, p. 157.

In chalk–dolerite contact-zone, Scawt Hill, Larne, Co. Antrim. C. E. Tilley, Geol. Mag., 1948, vol. 85, p. 216 [M.A. **10**–355]; Min. Mag., 1951, vol. 29, p. 644.

In metamorphosed limestone, Camas Mòr, Isle of Muck, Inverness-shire. C. E. Tilley, Bull. Comm. Géol. Finlande, 1947, no. 140, p. 99 [M.A. **10**–285]

Mossottite $(Ca,Sr)CO_3$ Orthorhomb

Variety of aragonite. Lunehead mine, Yorkshire. J. N. Friend an J. P. Allchin, Min. Mag., 1940, vol. 25, p. 591.

Nacrite $Al_2O_3.2SiO_2.2H_2O$ Monoclin

A kaolin mineral in syenite, Groby, Leicestershire. G. F. Claringbul Min. Mag., 1952, vol. 29, p. 973.

In metamorphosed lava, Shap, Westmorland. R. J. Firman, Mi Mag., 1953, vol. 30, p. 199.

[The 'nacrite' listed by Greg and Lettsom (1858) is muscovite.]

Nadorite $PbSbO_2Cl$ Orthorhomb

With jamesonite, Bodannon mine, St. Endellion, Cornwall. A Russell, Min. Mag., 1927, vol. 21, p. 272.

With jamesonite, Trevinnick mine, St. Kew, Cornwall. A. W. G Kingsbury, Min. Mag., 1957, vol. 31, p. 499.

Natrojarosite $NaFe_3(SO_4)_2(OH)_6$ Rhombohedra

Wanthwaite mine, St. John-in-the-Vale, Threlkeld, Cumberlan A. W. G. Kingsbury and J. Hartley, Min. Mag., 1958, vol. 31, p. 814.

Neptunite $(Fe,Mn)O.(Na,K)_2O.4SiO_2.TiO_2$ Monoclini

In quartz-syenite veins in limestone, Barnavave, Carlingford, Co Louth. S. R. Nockolds, Min. Mag., 1950, vol. 29, p. 27.

Omphacite $(Ca,Mg,Fe)SiO_3$ Monoclini

A pyroxene in eclogite, Glenelg, Inverness-shire. A. R. Alderma Quart. Journ. Geol. Soc. London, 1936, vol. 92, p. 493.

Orpiment As_2S_3 Monoclini

In clay, Clevedon, Somersetshire. A. Russell, Min. Mag., 1944, vo 27, p. 3. [An earlier record, A. Gages, 1860, from Silvermines, Co Tipperary, is doubted.]

Paracelsian $BaAl_2Si_2O_8$ Pseudo-orthorhombi

In metamorphosed manganese ore, Benallt mine, Rhiw, Carnarvon shire. L. J. Spencer, Min. Mag., 1942, vol. 26, p. 231.

Paratacamite $Cu_2Cl(OH)_3$ Rhombohedra

Botallack mine, St. Just, Cornwall. F. A. Bannister, M. H. Hey, an G. F. Claringbull, Min. Mag., 1950, vol. 29, p. 280.

Pennantite $(Mn,Al)_6(Si,Al)_4O_{10}(OH)_8$

A chloritic mineral in manganese ore, Benallt mine, Rhiw, Carnarvonshire. W. C. Smith, F. A. Bannister, and M. H. Hey, Min. Mag., 1946, vol. 27, p. 217.

Periclase MgO Cubic

In metamorphosed limestone, Carlingford, Co. Louth. G. D. Osborne, Geol. Mag., 1932, vol. 69, p. 214 [M.A. **5**–134]; S. R. Nockolds, Min. Mag., 1947, vol. 28, p. 157.

In dolomite–granite contact-zone, Broadford, Skye, Inverness-shire. C. E. Tilley, Geol. Mag., 1948, vol. 85, p. 216 [M.A. **10**–354]; Min. Mag., 1951, vol. 29, p. 634.

In metamorphosed limestone, Camas Mòr, Isle of Muck, Inverness-shire. Bull. Comm. Géol. Finlande, 1947, no. 140, p. 101 [M.A. **10**–285].

Picromerite $K_2Mg(SO_4)_2.6H_2O$ Monoclinic

In salt deposits, Whitby, Yorkshire. F. H. Stewart, Proc. Yorks. Geol. Soc., 1954, vol. 29, p. 228 [M.A. **12**–499].

Pigeonite $(Mg,Fe,Ca)SiO_3$ Monoclinic

In quartz–dolerite and tholeiite, central Scotland. F. Walker, Min. Mag., 1935, vol. 24, p. 141.

In olivine–dolerite, Portrush, Co. Antrim. N. Harris, Proc. Roy. Irish Acad., 1937, vol. 43, sect. B, p. 95 [M.A. **7**–25].

Pinnoite (?) $MgB_2O_4.3H_2O$ Tetragonal

In salt deposits, Sleights, Eskdale, Yorkshire. G. Armstrong *et al.*, Min. Mag., 1951, vol. 29, p. 672.

Pisanite $(Fe,Cu)SO_4.7H_2O$ Monoclinic

Parys Mountain, Anglesey, Carnarvonshire. L. Bor, Min. Mag., 1950, vol. 29, p. 63.

Plombierite $CaO.SiO_2.nH_2O$ Gelatinous

Chalk–dolerite contact-zone, Ballycraigy, Larne, Co. Antrim. J. D. C. McConnell, Min. Mag., 1954, vol. 30, p. 293; 1955, vol. 30, p. 672.

Polyhalite $K_2Ca_2Mg(SO_4)_4.2H_2O$ Monoclinic

In salt deposits, Eskdale, Yorkshire. C. E. Tilley, Min. Mag., 1943, vol. 26, proc. (for 1942) p. lvii; G. M. Lees and A. H. Taitt, Quart. Journ.

Geol. Soc. London, 1946, vol. 101 (for 1945), pp. 268, 271; F. H. Stewar
Min. Mag., 1949, vol. 28, pp. 631, 657; 1951, vol. 29, p. 449; G. Arm
strong *et al.*, Min. Mag., 1951, vol. 29, p. 667; L. R. Raymond, Quar
Journ. Geol. Soc. London, 1953, vol. 108 (for 1952), p. 284 [M.A. **12**
283].

Portlandite $Ca(OH)_2$ Hexagona

In chalk–dolerite contact-zone, Scawt Hill, Larne, Co. Antrim. C. F
Tilley, Min. Mag., 1933, vol. 23, p. 419.

Pumpellyite $Ca_4(Al,Mg,Fe)_6Si_6O_{23}(OH)_3.2H_2O$ Monoclini

In spilitic lava, Builth, Brecknockshire. G. D. Nicholls, Proc. Geo
Soc. London, 1957, session 1956–57, p. 8.

In Ordovician spilitic lava, Lendalfoot, south Ayrshire. T. W
Bloxam, Min. Mag., 1958, vol. 31, p. 811.

Pyrophanite $MnTiO_3$ Rhombohedra

In manganese ore, Benallt mine, Rhiw, Carnarvonshire. W. C
Smith, F. A. Bannister, and M. H. Hey, Min. Mag., 1946, vol. 27, p. 218
W. C. Smith and G. F. Claringbull, ibid., 1947, vol. 28, p. 108.

Pyroxmangite $(Fe,Mn)SiO_3$ Triclini

In grunerite–garnet schist, Glen Beag, Glenelg, Inverness-shire. C. F
Tilley, Amer. Min., 1937, vol. 22, p. 720 [M.A. **6**–528].

Rammelsbergite $NiAs_2$ Orthorhombi

Glencrieff mine, Wanlockhead, Dumfriesshire. A. K. Temple, Min
Mag., 1954, vol. 30, p. 541.

Rankinite $3CaO.2SiO_2$ Monoclini

In chalk-dolerite contact-zone, Scawt Hill, Larne, Co. Antrim. C. F
Tilley, Min. Mag., 1942, vol. 26, p. 190.

In metamorphosed limestone at gabbro contact, Camas Mòr, Isle o
Muck, Inverness-shire. C. E. Tilley, Bull. Comm. Géol. Finlande, 194
no. 140, p. 100 [M.A. **10**–285].

Rashleighite $Cu(Al,Fe)_6(PO_4)_4(OH)_8.5H_2O$ Triclin

Bunny mine, St. Austell, and Castle-an-Dinas mine, St. Columb Majo
Cornwall. A. Russell, Min. Mag., 1948, vol. 28, p. 353; 1952, vol. 2
p. 819. (*See* Turquoise.)

Realgar AsS Monoclinic

With native arsenic in dolerite (greenstone), Burraton Combe quarry, Saltash, Cornwall. A. Russell, Min. Mag., 1944, vol. 27, p. 3.

Rinneite $K_3NaFeCl_6$ Rhombohedral

In salt deposits, Aislaby, Eskdale, Yorkshire. F. H. Stewart, Min. Mag., 1949, vol. 28, p. 622; 1951, vol. 29, p. 563.

Rockbridgeite $(Fe'',Mn)Fe'''_4(PO_4)_3(OH)_5$ Orthorhombic

Previously confused with dufrenite. Cornwall and Devonshire. A. W. G. Kingsbury, Min. Mag., 1957, vol. 31, p. 429.

Römerite (?) $Fe''Fe'''_2(SO_4)_4.14H_2O$ Triclinic

Parys Mtn, Anglesey, Carnarvonshire. L. Bor, Min. Mag., 1950, vol. 29, p. 66.

Rosasite $(Cu,Zn)_2CO_3(OH)_2$ Botryoidal

Lake District, Cumberland (7 localities), and Cornwall. A. W. G. Kingsbury and J. Hartley, Min. Mag., 1957, vol. 31, p. 501.

Russellite $(Bi_2,W)O_3$ Tetragonal (?)

Castle-an-Dinas mine, St. Columb Major, Cornwall. M. H. Hey and F. A. Bannister, Min. Mag., 1938, vol. 25, p. 41; A. Russell, ibid., 1938, vol. 25, p. 49; 1944, vol. 27, p. 2. Cf. L. G. Sillén and K. Lundborg, Arkiv Kemi, Min. Geol., 1953, vol. 17A, no. 21 [M.A. **9**–99].

Rutherfordine UO_2CO_3 Orthorhombic

South Terras mine, St. Stephens, Cornwall. C. C. James, Trans. Roy. Geol. Soc. Cornwall, 1947, vol. 17 (pt. 5, for 1945), p. 257 [M.A. **10**–114]; J. Robson, ibid., 1949, vol. 17 (pt. 8, for 1948), p. 462 [M.A. **11**–273].

Sapphirine $(Mg,Fe)_2Al_4SiO_{10}$ (?) Monoclinic

In hornstone, Tievebulliagh, Co. Antrim. S. O. Agrell quoted by H. Sørenson, Meddel. om Grønland, 1955, vol. 137, no. 1, p. 25 [M.A. **13**–202].

Scawtite $Ca_7Si_6O_{16}CO_3(OH)_4$ Monoclinic

In chalk–dolerite contact-zone, Scawt Hill, Larne, Co. Antrim. C. E. Tilley, Nature, London, 1929, vol. 124, p. 896; Min. Mag., 1930, vol. 22, p. 222; ibid., 1938, vol. 25, p. 38.

In chalk–dolerite contact-zone, Ballycraigy, Larne, Co. Antrim J. D. C. McConnell, Min. Mag., 1954, vol. 30, p. 295.

Sepiolite $Mg_3Si_4O_{11}.5H_2O$ (?) Compac

In serpentinite, Lizard, Cornwall. S. Caillère, Bull. Soc. Franç. Min Crist., 1949, vol. 72, p. 5; S. Caillère and S. Hénin, Nature, London, 1949 vol. 163, p. 962 [M.A. **11**–50].

Siderophyllite $H_4K_2Fe_3Al_4Si_6O_{24}$ Monoclini

Variety of biotite, in greisen veins in granite, Mourne Mts., Co. Down S. R. Nockolds and J. E. Richey, Amer. Journ. Sci., 1939, vol. 237 p. 39 [M.A. **7**–305].

Sphaerodialogite Variety of rhodochrosite, $MnCO_3$ Rhombohedra

In manganese ore, Harlech, Merionethshire. A. W. Woodland, Quart Journ. Geol. Soc. London, 1939, vol. 95, p. 34 [M.A. **7**–437].

Spurrite $2Ca_2SiO_4.CaCO_3$ Monoclinic

In chalk–dolerite contact-zone, Scawt Hill, Larne, Co. Antrim. C. E Tilley, Min. Mag., 1929, vol. 22, p. 78; 1931, vol. 22, p. 443; 1938, vol 25, p. 38.

In limestone–gabbro contact-zone, Camas Mòr, Isle of Muck, Inverness-shire. C. E. Tilley, Bull. Comm. Géol. Finlande, 1947, no. 140 p. 100 [M.A. **10**–285].

In metamorphosed limestone, Carlingford, Co. Louth. S. R. Nockolds Min. Mag., 1947, vol. 28, p. 155.

Gabbro intrusion in limestone, Camphouse, Ardnamurchan, Argyllshire. S. O. Agrell, Min. Soc. Notice, 1950, no. 69 [Amer. Min., **38**–1080]

Stichtite $Mg_6Cr_2(OH)_{16}CO_3.4H_2O$ Rhombohedra

Replacing chromite in serpentinite, Cunningsburgh, Shetland Islands H. H. Read and B. E. Dixon, Min. Mag., 1933, vol. 23, p. 309. (*See* Barbertonite.)

Sylvine KCl Cubic

In salt deposits, Eskdale, Whitby, Yorkshire. G. M. Lees and A. H Taitt, Quart. Journ. Geol. Soc. London, 1946, vol. 101 (for 1945), p 268; F. H. Stewart, Min. Mag., 1949, vol. 28, p. 622; 1951, vol. 29, pp 451–563; G. Armstrong *et al.*, Min. Mag., 1951, vol. 29, p. 667; L. R Raymond, Quart. Journ. Geol. Soc. London, 1953, vol. 108 (for 1952) p. 288 [M.A. **12**–283].

Szájbelyite $2MgO.B_2O_3.H_2O$ Orthorhombic (?)

In chalk–dolerite contact-zone, Broadford, Skye, Inverness-shire. C. E. Tilley, Geol. Mag., 1948, vol. 85, p. 215 [M.A. **10**–355]; Min. Mag., 1951, vol. 29, p. 638.

Tephroite Mn_2SiO_4 Orthorhombic

In manganese ore, Benallt mine, Rhiw, Carnarvonshire. W. C. Smith, F. A. Bannister, and M. H. Hey, Min. Mag., 1944, vol. 27, p. 40.

In manganese ore, Treburland mine, Altarnun, Cornwall. A. Russell, Min. Mag., 1946, vol. 27, p. 224; C. E. Tilley, ibid., p. 236.

Thucholite Pitchblende+hydrocarbon

Laxey lead-mine, Isle of Man. C. F. Davidson and S. H. U. Bowie, Bull. Geol. Surv. Gt. Britain, 1951, no. 3, p. 4 [M.A. **11**–325].

Tilleyite $3CaO.2SiO_2.2CaCO_3$ Monoclinic

In metamorphosed limestone, Carlingford, Co. Louth. S. R. Nockolds, Min. Mag., 1947, vol. 28, p. 151; J. V. Smith, Acta Cryst., 1955, vol. 6, p. 9 [M.A. **12**–337].

In metamorphosed limestone, Camas Mòr, Isle of Muck, Inverness-shire. C. E. Tilley, Bull. Comm. Géol. Finlande, 1947, no. 140, p. 100 [M.A. **10**–285].

Turquoise $CuAl_6(PO_4)_4(OH)_8.5H_2O$ Triclinic

Castle-an-Dinas mine, St. Columb Major, and Bunny mine and Gunheath pit, St. Austell, Cornwall. A. Russell, Min. Mag., 1938, vol. 25, p. 54; 1952, vol. 29, pp. 819, 909. (*See* Rashleighite.)

Varlamoffite $H_2SnO_3+Fe_2O_3$, &c.

In tin-mines, Cligga mine, Perranzabuloe; Bunny mine, St. Austell; Kit Hill mine, Stokeclimsland, Cornwall. A. Russell and E. A. Vincent, Min. Mag., 1952, vol. 29, p. 817.

Veatchite $SrB_6O_{10}.2H_2O$ Monoclinic

In salt deposits, Aislaby, Eskdale, Yorkshire. F. H. Stewart, R. A. Chalmers, and R. Phillips, Min. Mag., 1954, vol. 30, p. 389.

Xanthophyllite $Ca(Mg,Al)_3(Al,Si)_4(O,OH)_{12}$ Monoclinic

In dolomite–granite contact-zone, Broadford, Skye, Inverness-shire. C. E. Tilley, Geol. Mag., 1948, vol. 85, p. 216 [M.A. **10**–354]; Min. Mag., 1951, vol. 29, pp. 625, 634.

Xonotlite $CaSiO_3.nH_2O$

In chalk–dolerite contact-zone, Scawt Hill, Larne, Co. Antrim. C. E Tilley, Min. Mag., 1931, vol. 22, p. 444.

In dolomite–granite contact-zone, Broadford, Skye, Inverness-shire C. E. Tilley, Geol. Mag., 1948, vol. 85, p. 216 [M.A. **10**–354]; Min. Mag. 1951, vol. 29, p. 634.

Zinckenite $PbSb_2S_4$ or $Pb_6Sb_{14}S_{27}$ Orthorhombic (pseudo hexagonal

In greisen, Grainsgill, Carrock Fell, Cumberland. A. W. G. Kingsbury and J. Hartley, Min. Mag., 1956, vol. 31, p. 297.

The Supplementary List of British Minerals (1898) and Second Supplementary List of British Minerals (1931), above referred to, are reprinted below by permission of the British Association [M. H. H.].

Supplementary List of British Minerals. [1898].

By L. J. Spencer, *M.A.*, *F.G.S.*

During the forty years which have elapsed since the publication o Greg and Lettsom's 'Manual of the Mineralogy of Great Britain and Ireland' a considerable number of species, variety, and other names have been added to the list of minerals occurring in the British Isles. In 185 Greg and Lettsom recognized 241 British species; but of these only 20 are given as numbered species by Dana in the sixth edition (1892) of hi 'System of Mineralogy'. To this list may now be added 84 more, bring ing the total number of British species up to 293, as compared with th total of 824 known mineral species recognized by Dana in 1892. Owin to the difficulty, in some cases, of defining a mineral species, to th uncertainty of some of the determinations, and to the fact that a sys tematic search through the whole of the literature has not yet bee made, these numbers can only be considered approximate.

Some of the most notable additions are of minerals which have bee detected by the microscopical examination of rock-sections, e.g. nephe ite, nosean, and various felspars, pyroxenes, and amphiboles.

In the following list are added 113 other names which have bee applied to British minerals; the species are distinguished by italics. Th first observer and date have been added (in parenthesis) wherever pos sible; in other cases the earliest reference found is given.

Abriachanite (Heddle, 1879).
Achroite (Collins, 1876).
Ægirite (Teall, 1888).
Aikinite (Museum Pract. Geol.).
Albertite (Morrison, 1884).
Alunogen (Smithe, 1882).
Amazon stone (Heddle, 1877).
Amblystegite (Judd, 1885).
Andesine (Heddle, 1877).
Andrewsite (Maskelyne, 1871).
Anorthite (Haughton, 1856).
Antigorite (Heddle, 1878).
Antimony? (Garby, 1848).
Atacamite (Church, 1865).
Balvraidite (Heddle, 1880).
Baricalcite (Breithaupt, 1841).
Barytocelestite (Collie, 1878).
Bastite.
Bathvillite (Williams, 1863).
Bauxite (Sutherland, 1870).
Bayldonite (Church, 1865).
Beekite.
Beraunite (Greg, 1860).
Bhreckite (Heddle, 1879).
Botallackite (Church, 1865).
Bowlingite (Hannay, 1877).
Braunite? (Collins, 1871).
Bruiachite (Macadam, 1886).
Bytownite (Teall, 1884).
Cantonite (Davies, 1877).
Cathkinite (Glen and Young, 1882).
Celadonite (Heddle, 1879).
Centrallassite (How, 1878).
Chalcosiderite (Maskelyne, 1875).
Chenevixite (Adam, 1866).
Chloritoid (Heddle, 1879).
Chloropal (Church, 1866).
Chlorophyllite (Heddle, 1882).
Chrome-diopside (Teall, 1888).
Chrysoberyl (Haughton, 1856).
Chrysotile.
Churchite (Church, 1865).
Cleavelandite (Heddle, 1877).
Clinochlore (British Museum).
Cloustonite (Heddle, 1880).
Coccolite (Heddle, 1877).
Collyrite (Gladstone, 1862).
Cotterite (Harkness, 1878).
Craigtonite (Heddle, 1882).
Crocidolite (Heddle, 1879).
Cryptolite (Church, 1872).
Danalite (Miers and Prior, 1892).
Daphnite (Tschermak, 1891).
Delessite (Heddle, 1879).
Demidoffite (Greg, 1860).
Descloizite (Frenzel, 1875).
Devilline (Pisani, 1864).
Dolianite (Des Cloizeaux, 1862).
Dudgeonite (Heddle, 1889).
Dufrenite (Kinch and Butler, 1886).
Duporthite (Collins, 1877).
Edenite (Heddle, 1878).
Electrum (Forbes, 1867).
Ellonite (Heddle, 1882).
Enstatite.
Enysite (Collins, 1876).
Eosite (Schrauf, 1871).
Eulytite (Collins, 1881).
Evansite (Woodward, 1884).
Ferrite (Heddle, 1882).
Fibrolite (Heddle, 1882).
Fichtelite (Macadam, 1889).
Freieslebenite? (Museum Pract. Geol.).
Funkite (Heddle, 1882).
Genthite (Heddle, 1878).
Gersdorffite (Forbes, 1868).
Gigantolite (Heddle, 1882).
Glaucophane (Blake, 1888).
Gramenite (Collins, 1877).
Grastite (Heddle, 1878).
Grossular (Heddle, 1878).
Grünlingite (Muthmann Schröder, 1897).
Halloysite (Heddle, 1882).
Haughtonite (Heddle, 1879).
Hausmannite (Goodchild, 1875).
Henwoodite (Collins, 1876).
Hibbertite (Heddle, 1878).
Hisingerite (Church, 1870).
Hovite (Gladstone, 1862).
Hullite (Hardman, 1878).
Hydrated labradorite (Heddle, 1880).
Hydrocerussite (Heddle, 1889).
Hydrophilite (Dana, 1892).
Hydroplumbite (Heddle, 1889).
Hydrozincite (Goodchild, 1883).
Iddingsite? (Arnold-Bemrose, 1894).
Igelströmite (Heddle, 1878).
Inverarite (Heddle, 1883).
Jarrowite (Lebour, 1887).
Johannite? (Garby, 1848).
Langite (Maskelyne, 1864).
Latrobite (Heddle, 1877).
Lepidomelane (Haughton, 1859).
Lettsomite (British Museum).
Leuchtenbergite (Glen and Young, 1876).

Leucoxene (Geikie, 1879).
Limnite (Church, 1865).
Linnœite (Terrill & Des Cloizeaux, 1880).
Liskeardite (Maskelyne, 1878).
Loganite (Harkness, 1866).
Löllingite (Collins, 1871).
Ludlamite (Field, 1877).
Lussatite (Mallard, 1890).
Lyellite (Maskelyne, 1864).
Marmolite (Heddle, 1878).
Martite (Heddle, 1882).
Massicot? (Collins, 1871).
Melanite (Teall, 1892).
Microcline.
Mirabilite (Glen and Young, 1876).
Monazite (Miers, 1885).
Montmorillonite (Collins, 1878).
Mottramite (Roscoe, 1876).
Mountain silk, &c. (Heddle, 1879).
Necronite (Heddle, 1877).
Neotype (Breithaupt, 1841).
Nephelite (Allport, 1871).
Nephrite (Heddle, 1878).
Nosean (Allport, 1874).
Okenite (Glen and Young, 1876).
Oligoclase (Haughton, 1862).
Omphacite (Teall, 1891).
Orangite (Heddle, 1883).
Ottrelite (Hutchings, 1889).
Pargasite (Heddle, 1878).
Penninite (Heddle, 1878).
Penwithite (Collins, 1878).
Perthite (Heddle, 1883).
Phlogopite? (Heddle, 1878).
Pickeringite (British Museum).
Picotite (Bonney, 1877).
Picrolite (Heddle, 1878).
Piedmontite (Dana, 1892).
Pihlite (Heddle, 1879).
Pilolite (Heddle, 1879).
Plumboaragonite (Collie, 1889).
Plumbogummite (Dana, 1850).
Plumbonacrite (Heddle, 1889).
Polybasite (Joy, 1860).
Polytelite (Forbes, 1867).
Prasilite (Thomson, 1840).
Protolithionite (Sandberger, 1885).
Proustite (British Museum).
Pseudo-hypersthene, &c. (Heddle, 1878).
Pseudophite (Heddle, 1879).
Pyroaurite (Heddle, 1878).
Pyrophyllite (Foster, 1876).
Pyrosclerite (Heddle, 1879).
Reichite (Breithaupt, 1865).
Restormelite (Church, 1870).
Rhabdophane (Lettsom, 1878).
Riebeckite (Harker, 1888).
Rock silk, &c. (Heddle, 1879).
Rubislite (Heddle, 1879).
Sanidine.
Scapolite (Ormerod, 1869).
Schraufite? (Thomson, 1887).
Schrötterite (Dana, 1868).
Senarmontite (Davies, 1867).
Sericite.
Spangolite (Miers, 1893).
Spessartite (Heddle, 1878).
Stephanite (Davies, 1866).
Tallingite (Church, 1865).
Tavistockite (Church, 1865).
Thorite (Heddle, 1883).
Tobermorite (Heddle, 1880).
Totaigite (Heddle, 1878).
Tridymite (Lasaulx, 1876).
Turgite (Heddle, 1882).
Tyreeite (Heddle, 1881).
Uigite (Heddle, 1856).
Uralite.
Valentinite (Hall, 1868).
Vauquelinite (Davies, 1877).
Vermiculite (Parke, 1877).
Voltzite (Dana, 1868).
Walkerite (Heddle, 1880).
Waringtonite (Maskelyne, 1864).
Wicklowite (D'Achiardi, 1883).
Willemite (Glen and Young, 1876).
Wittichenite? (Collins, 1871).
Woodwardite (Church, 1866).
Xantholite (Heddle, 1879).
Xanthosiderite (Haughton, 1866).
Xonaltite (Heddle, 1882).
Zeunerite (Weisbach, 1872).

By L. J. Spencer, F.R.S.—*Second Supplementary List of British Minerals.* [1931].

The first of these lists was presented at the Bristol meeting in 1898. They are intended to supplement the still standard work of Greg and Lettsom's 'Mineralogy of Great Britain and Ireland' (1858). The present list includes 90 names, of which 46 (printed in italics) may be regarded as well-defined species with satisfactory records. This brings the total number of mineral-species that have been found in the British Isles up to 339, representing about one-third of the number of known species.

Aegirine-augite (Tyrrell, 1909).
Aenigmatite (Heddle, 1901).
Afwillite (Tilley, 1930).
Altaite (Des Cloizeaux, 1893).
Anorthoclase (Heddle, 1901).
Arfvedsonite (Seymour, 1900).
Barkevikite (Busz, 1900).
Bassetite (Hallimond, 1915).
Bertrandite (Bowman, 1911).
Botryolite (McLintock, 1910).
Calciopalygorskite (Fersman, 1908).
Carminite (Russell, 1910).
Cebollite (Tilley, 1931).
Celsian (Russell, 1911).
Chamosite (Rudler, 1905).
Chloanthite (Russell, 1922).
Chloroxiphite (Spencer, 1923).
Chonicrite (Heddle, 1901).
Christophite (Collins, 1879).
Claudetite (Hintze, 1904).
Clinozoisite (Thomson, 1908).
Collieite (Brown, 1927).
Cotunnite (Russell, 1920).
Crednerite (Spencer, 1923).
Crocalite (Heddle, 1901).
Crocoite (Brown, 1927).
Cummingtonite (Teall, 1907).
Dechenite (?) (Heddle, 1901).
Diabantite (Garnett, 1923).
Diaboleite (Spencer, 1923).
Diamond (?) (Heddle, 1901).
Diaspore (Tilley, 1927).
Diatomite (Macadam, 1884).
Dopplerite (Moss, 1903).
Dumortierite (Mackie, 1924).
Dundasite (Prior, 1906).
Embolite (Prior & Spencer, 1902).
Epichlorite (Garnett, 1923).
Ettringite (Tilley, 1931).
Forsterite (Clough & Pollard, 1899).
Fuchsite (Heddle, 1901).
Geocronite (Prior, 1902).
Gibbsite (British Museum).
Gilpinite (Larsen & Brown, 1917).
Glockerite (Dana, 1899).
Hastingsite (Tilley, 1931).
Hercynite (Teall, 1897).
Hitchcockite (Miers & Hartley, 1900).
Hydronephelite (Tilley, 1931).
Keilhauite (?) (Heddle, 1901).
Kilmacooite (Tichborne, 1885).
Larnite (Tilley, 1929).
Laurionite (Russell & Hutchinson, 1922).
Lepidocrocite (Heddle, 1901).
Magnesite (Heddle, 1901).
Magnesium-diopside (Rosenbusch, 1905).
Marmatite (Rudler, 1905).
Melilite (Flett, 1900).
Merwinite (Tilley, 1927).
Meta-torbernite (Hallimond, 1920).
Morenosite (Heddle, 1901).
Mullite (Bowen, Grieg, & Zies, 1924).
Nadorite (Russell, 1923).
Nigrine (Reynolds, 1871).
Orpiment (Gages, 1860).
Paralaurionite (Russell & Hutchinson, 1922).
Passyite (Rudler, 1905).
Perovskite (Elsden, 1904).
Petalite (McLintock, 1923).
Phenakite (Russell, 1911).
Pimelite (?) (Heddle, 1901).
Prochlorite (Heddle, 1892).
Ptilolite (Bøggild, 1922).
Rammelsbergite (?) (Heddle, 1901).
Scawtite (Tilley, 1929).
Semseyite (Smith, 1919).

Serpierite (Russell, 1927).
Sideroplesite (Heddle, 1901).
Spurrite (Tilley, 1929).
Stilpnomelane (Hallimond, 1924).
Stokesite (Hutchinson, 1899).
Titanaugite (Tyrrell, 1909).
Tschermigite (Shand, 1910).
Ullmannite (Spencer, 1910).
Uranospathite (Hallimond, 1915).
Urpethite (Dana, 1868).
Wurtzite (Noelting, 1887).
Yttrotantalite (?) (Heddle, 1901).
Xanthochroite (Rogers, 1917).
Xenotime (Gilligan, 1920).

SHORT COMMUNICATIONS

Pumpellyite from south Ayrshire.

Pumpellyite occurs in some of the altered Ordovician spilitic lavas that form part of the Girvan–Ballantrae complex. The mineral is particularly abundant in lavas exposed near outcrops of glaucophane-schist and serpentinite at Knockormal Farm, 1 mile east of Lendalfoot. Although originally described from copper deposits of the Keewanawan (Palache and Vasser, 1925), pumpellyite is probably best known from its occurrence in glaucophane-schists and associated rocks in California, Japan, Celebes, and elsewhere (Irving, Vonsen, and Gonyer, 1932; Switzer, 1950; Tsuboi, 1936; de Roever, 1947). The geological and petrological setting of the Girvan–Ballantrae complex is very similar to that in California, comprising metamorphic rocks of the greenschist type associated with spilitic lavas and basic and ultra-basic intrusives.

The lavas containing pumpellyite are dark green in colour, very fine-grained, and frequently porphyritic. The specimen from which the pumpellyite was separated had a specific gravity of 2·97. The pumpellyite occurs mainly in quartz-veins together with varying amounts of albite and calcite. In most cases the veins occupy shear-planes traversing the rock. Unsheared parts of the lava exhibit a typical variolitic or subvariolitic texture with turbid microlitic plagioclase set in a matrix consisting largely of brown glass charged with abundant iron-ore. The plagioclase is frequently sub-isotropic and altered to dense aggregates of calcite, quartz, albite, pumpellyite, and indeterminate brown sub-opaque material.

Pumpellyite is generally developed along the walls of the quartz-veins and forms needle-like clusters with a stellate habit (fig. 1). The crystals are elongated parallel to *b* which is also the direction of maximum absorption (β), and the trace of a strong {001} cleavage. When calcite forms a major constituent of the veins the pumpellyite assumes a 'chloritic' habit composed of a dense matt of variously orientated fibres (Hutton, 1937, p. 531). This habit is also characteristic of pumpellyite occurring in many irregular patches throughout the remainder of the rock, including prominent calcite–pumpellyite pseudomorphs after plagioclase phenocrysts. Minor amounts of epidote may accompany the pumpellyite in the calcite veins. The absence of any marked thermal

alteration in the lava indicates that pumpellyite formed at low tem peratures in association with processes bringing about albitization of the plagioclase.

Chemical and optical analyses. About 200 mg. of pumpellyite was separated for analysis; the powder is pale yellow with a density of 3·23

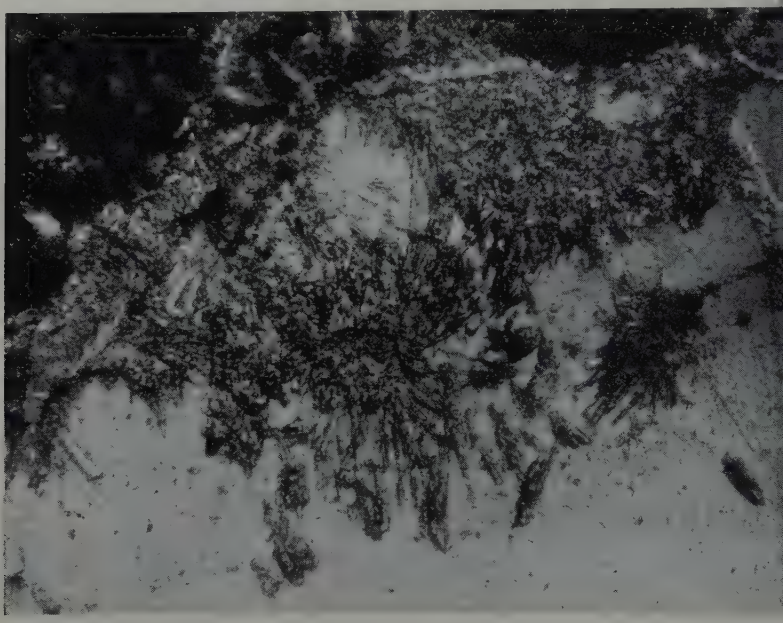

FIG. 1. Needles of pumpellyite showing stellate habit in quartz-vein. Ordinary light, ×90.

The analysis gave: SiO_2 37·20 %, TiO_2 0·18, Al_2O_3 24·84, Fe_2O_3 2·02 FeO 3·11, MnO 0·12, MgO 2·70, CaO 22·91, Na_2O 0·22, K_2O 0·00, H_2O+ 6·60, sum 99·90 %. Sp. gr. 3·23. Recalculated to metal atoms per hal unit cell this gives: Si 6·03, Ti 0·03, Al 4·72, Fe‴ 0·23, Fe″ 0·33, Mn 0·02 Mg 0·65, Ca 3·90, Na 0·06, OH − 3·00, H_2O 2·03; Al+Ti+Fe‴ 4·98 Mg+Fe″+Mn 1·00; Ca+Na 3·96; which shows good correspondenc with the proposed formula for pumpellyite: $Ca_4R_6Si_6O_{23}(OH)_3.2H_2O$ where R = (Ti,Al,Fe‴,Fe″,Mn,Mg) and (Ti+Al+Fe‴) : (Fe″+Mn+ Mg) = 5:1 (Irving, Vonsen, and Gonyer, 1932; Coombs, 1953). Th optical properties of the mineral are: α 1·691, colourless; β 1·693, blue green; γ 1·701, colourless; $\gamma-\alpha$ 0·010; $2V\gamma$ 12 to 25° (strong dispersion) $\alpha:a$ 26° ($\beta = b$). Owing to the strong dispersion, values for optic axia

angle and extinction angle are approximate determinations in sodium-light.

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D. S. Coombs, Min. Mag., 1953, vol. 30, p. 113.
W. P. de Roever, Geol. Expl. Celebes. Amsterdam, 1947 [M.A. **11**-200].
C. O. Hutton, Min. Mag., 1937, vol. 24, p. 529.
J. Irving, M. Vonsen, and F. A. Gonyer, Amer. Min., 1932, vol. 17, p. 338 [M.A. **5**-233].
C. Palache and H. E. Vasser, Amer. Min., 1925, vol. 10, p. 412 [M.A. **3**-8].
G. Switzer, Calif. Div. of Mines, Bulletin 161, p. 51, 1950.
S. Tsuboi, Japan. Journ. Geol. Geogr., 1936, vol. 13, p. 333 [M.A. **7**-30].

Jarosite and natrojarosite from the Lake District.

Jarosite ($KFe_3(SO_4)_2(OH)_6$) is a mineral that may easily be overlooked, commonly occurring as soft earthy aggregates or crusts of microscopic crystals, ranging from buff- and golden-yellow to brown in colour; in these forms it can readily be confused with 'limonitic' matter. While it does not appear to have been previously recorded in Britain, we have found that it is by no means rare in the Lake District and we have confirmed it from the following localities:

Grains Gill, Carrock Fell: crusts of very small golden-yellow to brown crystals on white vein-quartz from the 'granitic' suite of veins; it is associated with pale yellowish-buff, fine-grained, earthy ferriferous alunite (B.M. 1956, 76; 1958, 43).

Higher Brandy Gill, Carrock Fell: small compact masses of glistening yellow-brown crystals in the outcrop of an east–west lead–copper vein near the head of the gill on the western side (B.M. 1958, 44); also in similar crystalline patches in vein-quartz derived from an old cross-cut on the eastern side.

Burdell Gill, Caldbeck Fells: in gossany vein-stuff from a north–south iron–manganese vein tried near the head of the Gill, 400 yards south-west of Coomb Height; forms compact brown crystalline masses and small (up to 2 mm.) brown rhombohedral crystals on goethite and saccharoidal quartz. The crystals resemble brownish pharmacosiderite also occurring here (B.M. 1956, 78; 1958, 45).

Netherrow Brow, Caldbeck: in oxidized vein-material derived from an old collapsed cross-cut near the farm of Nether Row, driven on a north-west–south-east vein, as crystalline aggregates, of a golden-brown colour, in saccharoidal quartz (B.M. 1956, 75; 1958, 46); also in similar vein-

material from an old cross-cut, almost certainly to the same vein between Sandbed and Potts Gill mines, half a mile to the south-east.

Potts Gill mine, Caldbeck: occurs as golden-yellow to brown fine-grained crystalline aggregates in oxidized vein-material from a small copper vein cut by the old no. 1 cross-cut; it is associated with various secondary arsenates.

Ingray Gill, Caldbeck: in oxidized vein-material from the northern most of the two old cross-cuts near the head of the gill; occurs in small brown rhombohedral crystals, some of them altering to 'limonite', with arsenates.

Higher Roughtongill, Caldbeck Fells: occurs as a yellow to brown powdery aggregate, associated with beaverite, beudantite, and chrysocolla in chalcedonic quartz, in the outcrop of the south vein along the flank of Balliway Rigg (B.M. 1956, 77; 1958, 47).

Wanthwaite mine, St. John-in-the-Vale, near Threlkeld: occurs as a coating of yellow, microscopic crystals on quartz, with pyrite, arsenopyrite, and graphite, from the southernmost of the two veins at this mine. This vein occupies a fault between black Skiddaw Slates (on the north) and Borrowdale Volcanics (B.M. 1958, 48).

Natrojarosite.

While the southernmost vein at the Wanthwaite mine yielded jarosite, the northern vein yielded its rarer relative, natrojarosite ($NaFe_3(SO_4)_2(OH)_6$). Practically no details are known about this mine, which included two veins about 200 yards apart; these veins have been referred to by J. Clifton Ward[1] and in the subsequent literature as 'copper-veins', but in fact the metallic minerals are mainly arsenopyrite and pyrite with some blende, galena, stibnite, and sulpho-salts, in a quartz–carbonate gangue; copper minerals are only present in traces. The northern vein occupies the faulted boundary between the main, eastern exposure of the St. Johns or Threlkeld microgranite (on the north) and Skiddaw Slates, and has been tried from three cross-cuts just beneath its outcrop on Clough Head, from another cross-cut some 300 feet lower down, and from a day-level, directly on the vein, a further 100 feet below the middle cross-cut. The vein is extremely gossany at its outcrop and it was in such material that the natrojarosite was found, in aggregates of small (up to 0·5 mm.) golden-brown rhombohedral crystals on massive and botryoidal goethite, the crystals showing combinations of the basal plane *c* {0001} with *e* {10$\bar{1}$1} and *s* {01$\bar{1}$2}. In the hand-specimen they

are indistinguishable from jarosite but were confirmed as natrojarosite by their X-ray powder patterns (B.M. 1958, 49).

A closely allied mineral, containing 1·99 % Na_2O and 3·5 % K_2O and showing affinities to both natrojarosite and alunite, has been collected from a quartz–carbonate vein cutting a highly altered quartz-dolerite at Embleton Quarry, about 2 miles west of Bassenthwaite Lake railway station.

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Department of Geology, University of Leeds. J. HARTLEY

[1] The Geology of the northern part of the English Lake District, 1876 (Mem. Geol. Survey).

Two beryllium minerals new to Britain: euclase and herderite.

BERYLLIUM minerals are by no means common in the west of England, and many of the recorded occurrences are from old specimens or localities where they are now no longer obtainable or extremely rare. Several further new occurrences of beryl and other rarer beryllium minerals have, however, recently come to light in both Devon and Cornwall; among these rarer species, euclase and herderite are interesting additions to the list.

Euclase. Cligga Head has long been known as a locality for well-crystallized topaz; though widely distributed as an accessory in the greisen and as small crystals in many of the quartz-veins and joints, the best specimens have been found in quartz–tourmaline–topaz veins, which are barren of metallic minerals; these veins are somewhat restricted in their occurrence. The crystals are prismatic in habit, usually slightly flattened laterally in one direction, and are colourless, yellowish, or sometimes bluish, and may reach 7 or 8 mm. in length. A description of the occurrence and of the crystals found there was given by Sir Arthur Russell[1] in 1924. Though now not so easily found as previously, crystals of topaz can still be collected by breaking open portions of quartz-veins, *in situ* when accessible, or in blocks of greisen lying among the mining debris or which may have fallen from the cliffs.

It is in part of a quartz-vein from one of such blocks, collected in 1949, that euclase has now been found. The block was among a pile of

similar material covering a small shaft in the wolfram-workings and though fresh and evidently taken out fairly recently, its exact origin is unfortunately not known. In the course of breaking up this piece of matrix, with other similar material, a pale bluish prismatic crystal became detached and fell out of a shattered cavity; this crystal was about 8 mm. long and at first sight might have been taken for a typical bluish topaz crystal from this locality. The most unusual, and at once noticeable feature, however, was that it showed a very distinct, longitudinal, prismatic cleavage and not the characteristic basal cleavage of topaz. Fortunately it was possible to pick out and recover from the broken vein-material a number of fragments of the remainder of the crystal, spectrograms of which, taken by S. Ross Taylor in this Department, showed that the mineral contained beryllium. The cleavage suggested that the crystal might be euclase and the spectrographic examination provided additional and stronger evidence: a subsequent X-ray powder photograph has confirmed the identification.

Only this one crystal (B.M. 1958, 40) was found, in a not very large piece of the matrix that appeared to have come from a vein barren of metallic minerals: I have found small crystals of topaz in parts of other veins, collected in recent years on different occasions, and it seems probable that the euclase is very restricted in its occurrence. It is possible that other specimens might be found by careful searching at Cligga Head.

Herderite. This rare calcium-beryllium phosphate was originally described from the tin-mines at Ehrenfriedersdorf in Saxony, a few specimens only having been found there prior to 1825. Herderite has since been found in pegmatite in Bavaria, at Mursinsk in the Urals, and at a number of New England localities in the U.S.A.; at all of these latter localities it occurs as a later-stage, hydrothermal mineral.

In 1920 Sir Arthur Russell[2] recorded the finding by himself at Wheal Cock, St. Just, Cornwall, of phenakite and scheelite; these minerals occurred in a matrix of loosely coherent, scaly, brownish-green, partially altered chlorite forming a loose block among other blocks of lode-material lying in the rocky inlet known as Stamps and Jowl Zawn,[3] at the foot of Roscommon Cliff, under Wheal Cock, one of the adits of which opens into the Zawn. In 1949, while on a visit to the St. Just area, I collected several specimens of almost identical scaly chlorite from the waste material lying round one of the old shafts of Wheal Cock just above and inland from the same Zawn, my attention having been attracted by some small buffish prisms of apatite embedded in part of it. In one of the specimens, which was friable and crumbly, there were

embedded several rather shattered, short prismatic crystals of phenakite, and some more buffish, apparently hexagonal crystals, which appeared to be apatite. These latter crystals were corroded and again much shattered and mostly came out in fragments, but on being powder-photographed, some of them gave an entirely different pattern; spectrograms were then taken which showed the presence of calcium and beryllium, and the mineral was confirmed as herderite (B.M. 1958, 41).

Following on this, I re-examined a number of other specimens of apatite I had collected on various occasions in Cornwall and, as a result, have found a second occurrence of herderite, at Colcerrow Quarry, Luxullyan. This and other adjoining quarries are well known for their specimens of fluorite, gilbertite, and, at Colcerrow in particular, apatite. The best-known apatites from here are pale blue in colour and from the development of many faces in the pyramid zones have a rather rounded appearance and habit. Other crystals are, however, sometimes more prismatic in habit and show other shades of colour, from colourless to whitish, yellowish, and greenish. Since the quarries ceased working, good specimens have been difficult to obtain, but in 1931 I was lucky in finding a few small specimens of crystallized apatite in vugs in some of the coarser pegmatitic granite at Colcerrow. The apatite crystals are generally implanted in or associated with globular aggregates of gilbertite on creamy-coloured crystals of felspar and may be also associated with albite, tourmaline, and somewhat smoky quartz. Most of them were colourless or very pale yellowish and showed the usual rather rounded form; on examination, however, some of these recently have proved to be herderite (B.M. 1958, 42), again showing the less common pseudo-hexagonal habit of the St. Just crystals, and being only with difficulty distinguishable from apatite.

It is possible that other occurrences of herderite may be found in pegmatite druses where there has been late-stage hydrothermal mineralization or that they have been overlooked or mistaken for apatite. In view of the scarcity of beryllium minerals in Cornwall and in Britain generally, the identification of herderite makes an interesting addition to the list.

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[1] Min. Mag., 1924, vol. 20, p. 225. [2] Ibid., 1920, vol. 19, p. 19.

[3] A Zawn in the St. Just area is a sheer steep-sided chasm in the cliffs formed by the walls of an outcropping mineral-vein, when the vein itself has disintegrated and fallen into the sea.

On carphosiderite.

Some labelled or previously reported 'carphosiderites' from St-Léger Mâcon, France, and from Greenland have been identified, on further examination, as jarosite[1, 2, 3] or as natrojarosite.[2] X-ray diffraction data on 'carphosiderite' from Greenland (U.S. National Museum, specimen number R 6266) have been published,[4, 5] but unfortunately were not accompanied by a chemical analysis. The studies on this mineral species are so scanty that it seems useful, in order to obtain more information on its validity or to avoid confusion, to check the chemical composition. The U.S. National Museum supplied for this examination a small sample of the 'carphosiderite' (U.S.N.M. R 6266) from Greenland and also of the natrojarosite[6] (U.S.N.M. 86932) from Soda Springs Valley, Nevada.

It will be seen that the 'carphosiderite' (U.S.N.M. R 6266) is in fact natrojarosite.

The X-ray powder photographs of both samples are almost identical. The chemical analysis of the 'carphosiderite', carried out on 0·33 g, gave the following figures:

	%	Recalculated %	Molecular ratio
Insoluble (hot HCl 1 : 1)	12·3	—	—
H_2O^- (< 110°)	1·3	—	—
H_2O^+ (> 110°)	12·1	13·9	0·771
SO_3	27·3	31·4	0·392
Fe_2O_3	42·7	49·1	0·307
$Al_2O_3+P_2O_5$	nil	—	—
CaO	nil	—	—
MgO	nil	—	—
Na_2O	4·8	5·5	0·089
K_2O	(0·1)	(0·1)	(0·001)
Total	100·6	100·0	—

In the insoluble were recognized some grains of a green monoclinic pyroxene, presumably augite, and of pyrite (both minerals checked by X-rays). The total water content was determined according to the method of Penfield as modified by Hartwig-Bendig.[7] The other components were estimated in the soluble fraction. Iron was determined by cerimetry. The calcined ammonia precipitate agreed with the Fe_2O_3 so determined, and from this the absence of weighable Al_2O_3 and P_2O_5 was concluded. The sulphate was weighed as $BaSO_4$ after removal of iron. The alkalis were determined by flame spectrophotometry using standards of similar base composition. The figure for potassium is un

certain, but indicates a maximum value, as this was the limit of sensitivity of the method.

The molecular ratios are in good agreement, for sodium, iron, and sulphur, with those of natrojarosite $NaFe_3(SO_4)_2(OH)_6$. The determined water content is, with respect to this formula, somewhat high, but it may be supposed that the material responsible for the insoluble slightly affects the water ratio.

My thanks are due to Dr. G. Switzer, Acting Curator, Division of Mineralogy and Petrology, U.S. National Museum, who kindly supplied the material.

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[1] A. Lacroix, Min. France, 1910, vol. 4, p. 145.
[2] R. Van Tassel, Bull. Inst. roy. Sci. nat. Belgique, 1956, vol. 32, no. 24, p. 9 [M.A. 13–369].
[3] A. A. Moss, Min. Mag., 1957, vol. 31, p. 407.
[4] S. B. Hendricks, Amer. Min., 1937, vol. 22, p. 773.
[5] C. M. Warshaw, ibid., 1956, vol. 41, p. 288.
[6] W. F. Hillebrand and S. L. Penfield, Amer. Journ. Sci., 1902, ser. 4, vol. 14, p. 211.
[7] M. Hartwig-Bendig, Zeits. angew. Min., 1941, vol. 3, p. 195.

Isokite from Mangualde, Portugal.

D. J. Fisher's discovery of isokite replacing triplite from the old tin mines of Slavkov, Bohemia,[1] an environment quite different from the carbonatite in which isokite was originally found,[2] aroused the writer's interest in other triplite localities. This has led to the identification as isokite of a mineral described but not named by A. M. de Jesus[3] from Mangualde, in the province of Beira, Portugal, which historically would now appear to be the first locality for this mineral. The mineral was described as a fluophosphate of calcium and magnesium, but was not named as the chemical analysis was unsatisfactory, totalling only 90·58 % (after the oxygen-fluorine correction), and it could only be compared with cryphiolite, the one substance then recorded of similar composition, but now a discredited species.[4]

As described by de Jesus, the mineral forms dull white earthy crusts on partially oxidized and hydrated triplite (his *metatriplite*). A specimen recently obtained through the courtesy of Professor de Jesus and J. Lima-de-Faria consists of massive black triplite (enclosing patches of pale brown triplite) thinly encrusted here and there in irregular cavities

with dull white earthy material. Optically this white material, as far can be determined, resembles isokite, being minutely crystalline aggregates of fibres or radiating platelets with negative elongation, near 1·590 and γ near 1·610. These figures are only approximate, a unfortunately the platelets are too small for determination of sign a optic axial angle. An X-ray powder photograph was taken by D J. D. C. McConnell who states that it is identical with his photograph the type isokite from Rhodesia.

As the main difference between isokite and de Jesus's analysis lies the phosphate content, the latter was redetermined on a small samp (5 mg.) freed of a little contaminating triplite, quartz, and felspar b means of the electromagnet and the centrifuge. The determinatio made by my colleague R. Pickup, revealed 37·0 % P_2O_5, in place 26·59 % reported by de Jesus. With this correction the analysis may b restated as CaO 31·52, MgO 20·69, P_2O_5 37·0, F 7·75, SiO_2 4·13, Fe 1·81, MnO 0·57, Na_2O 0·37, K_2O 0·25, H_2O- 0·16 %. The deficiency in t total has been eliminated and, allowing for some impurities, the co position is essentially that of isokite, $CaMgPO_4F$, and the interpretatio as near $Ca_3Mg_3(PO_4)_2O_2F_2$ can no longer stand.

At both Mangualde and Slavkov triplite is replaced by isokite, in plying substitution of the cations iron and manganese by calcium an magnesium, whereas at Isoka the isokite developed from dolomit the cations remaining passive while fluophosphate replaced carbonat According to de Jesus, but accepting later corrections to his nomencl ture, the phosphate sequence at Mangualde comprised a main hydr thermal phase of lithiophilite and triplite, with rare manganapatite, an a later mixed hydrothermal and supergene phase of heterosite, hureau ite, strengite, isokite, and torbernite. The record[6] of 'a mineral perha identical with wagnerite' from Mangualde evidently refers to isokite.

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[1] Min. Mag., 1957, vol. 31, p. 587.
[2] Ibid., 1955, vol. 30, p. 687.
[3] Comun. Serv. Geol. Portugal, 1936, vol. 19, pp. 154–156 [M.A. 6–440 (ana XIV)].
[4] Min. Mag., 1956, vol. 31, p. 346.
[5] M. H. Hey, An Index of Mineral Species, 2nd edn, London, 1955, item 22.1.1 p. 262.
[6] Dana's System of Mineralogy, 7th edn, vol. 2, 1951, p. 847.

PROCEEDINGS

OF THE

MINERALOGICAL SOCIETY

(1957)

EXCERPT MINUTES OF COUNCIL MEETING

Held in the Apartments of the Geological Society, Burlington House, London, W., 24 January 1957, at 3.15 p.m.

Prof. L. Hawkes, President, in the Chair.

THE following were provisionally elected Ordinary Members of the Society: Paul Bartholomé, Tom Neville Clifford, Knut Sigurdsön Heier, Vincent George Hill, Richard St. John Lambert, Charles Geoffrey Nicholson, Brian Simpson, Zaki Mohamed Zaghloul.

The deaths of Prof. Edward de Courcy Clarke, Dr. Frederick Arthur Henson, and Prof. Arthur Leonard Parsons were reported.

The resignations of Prof. P. G. H. Boswell, Mr. P. G. Cooray, Mr. D. B. Honeyborne, Mrs. J. M. Beck, and Mr. H. S. Peiser were accepted.

Reports were received from the Treasurer, the Editors, the Clay Minerals Group, and the Editorial Committee on Mineralogical Abstracts.

The publication of the Mineralogical Magazine by the Society and the direct sale of all publications from the Society's office were reported.

The nomination of a representative, when requested, to the Committee of the 5th Congress of the International Union of Crystallography, to be held in Cambridge, 1960, was agreed.

GENERAL MEETING

Held in the Apartments of the Geological Society, Burlington House, London, W., 24 January 1957, at 5 p.m.

Prof. L. Hawkes, President, in the Chair.

Forty Members and four Visitors were present.

The elections of eight Ordinary Members of the Society provisionally made by the Council were confirmed.

The following papers were read and discussed:

1. The nature of carphosiderite and allied basic sulphates of iron: by Dr. A. A. Moss (vol. 31, p. 407).

2. The illite in Old Red Sandstone soils and sediments: by Dr. R. C. Mackenzie (vol. 31, p. 681).

3. The paragenesis of garnet in charnockite, enderbite, and related granulites: by Dr. R. A. Howie and Dr. A. P. Subramaniam (vol. 31, p. 565).

4. Accurate determination of the cell dimensions of magnetite: by Dr. E. Z. Basta (vol. 31, p. 431).

5. Titanomagnetites from a differentiated teschenite sill: by Dr. J. F. G. Wilkinson (vol. 31, p. 443).

The following papers were taken as read:

1. An unusual cassiterite paragenesis and its genetic implications: by Dr. L. J. Lawrence (vol. 31, p. 402).

2. The normal-pattern finder: an aid to stereographic projection: by Mr. G. Boom (communicated by Dr. M. H. Hey) (vol. 31, p. 358).

3. Abraded pyrite crystals from the Witwatersrand gold mines: by Dr. J. J. Frankel (vol. 31, p. 392).

4. Dickite from sandstones in northern England and North Wales: by Dr. F. Smithson and Mr. G. Brown (vol. 31, p. 381).

5. Pyrosmalite in the Broken Hill lode, N.S.W.: by Dr. F. L. Stillwell and Dr. J. McAndrew (vol. 31, p. 371).

6. Heating experiments on amazonite: by Prof. Ivar Oftedal (vol. 31, p. 417).

7. The morphology of the Unter-Rupsroth montmorillonite: by Mr. H. L. Nixon and Mr. A. H. Weir (vol. 31, p. 413).

EXCERPT MINUTES OF COUNCIL MEETING

Held in the Apartments of the Geological Society, Burlington House, London, W., 28 March 1957, at 3.15 p.m.

Prof. L. Hawkes, President, in the Chair.

The following were provisionally elected Ordinary Members of the Society: David Murray Boyd, Fred A. Hildebrand, Juan Luis Martin-Vivaldi, Bryan Oliver Payne, William Antony Swithin Sarjeant, Brian John Skinner, John Bucknall Wright.

The deaths of Dr. Paul Grodzinski, Dr. Robert Campbell, and Prof. Frantisek Slavík were reported.

The resignation of Prof. H. Deuel was accepted.

The names of Mr. R. S. Long, Prof. S. B. McCaleb, Mr. A. J. C. Rodrigues, Mr. K. E. Seal, Mr. A. L. Silfverberg, and Dr. H. Towers were removed from the list of members for non-payment of subscriptions.

Reports were received from the Treasurer and the Editors, and from the Clay Minerals Group. The Treasurer presented the audited accounts for 1956, which were approved (vol. 31, pp. lx–lxiii).

Exemption from rates on the offices at 41 Queen's Gate, London, S.W. 7, had been claimed under the Scientific Societies Act, 1843.

Nominations for election to the Council were considered. The following were due to retire: President, Prof. L. Hawkes; Ordinary Members of Council, Mr. L. A. A. Thomas, Dr. M. K. Wells, Mr. S. H. U. Bowie, and Mr. B. W. Anderson. The following were proposed for election at the Anniversary Meeting in November 1957: President, Prof. C. E. Tilley; Ordinary Members of Council, Dr. R. J. W. McLaughlin, Dr. H. I. Drever, Mr. P. G. Embrey, and Dr. G. M. Brown.

The resignation of Miss M. S. Stapleton, Publications Clerk, was reported.

GENERAL MEETING

Held in the Apartments of the Geological Society, Burlington House, London W., 28 March 1957, at 5 p.m.

Prof. L. Hawkes, President, in the Chair.

Thirty-three Members and four Visitors were present.

The elections of seven Ordinary Members of the Society provisionally made by the Council were confirmed.

The following papers were read and discussed:

1. The photoelectric measurement of reflectivity: by Mr. S. H. U. Bowie (read by Mr. J. E. T. Horne) (vol. 31, p. 476).

2. Direct measurement of standard reflectances with the microphotometer: by Dr. A. F. Hallimond (vol. 31, p. 487).

3. Pyroxenes from the early and middle stages of fractionation of the Skaergaard intrusion, East Greenland: by Dr. G. M. Brown, with chemical analyses by Dr. E. A. Vincent and Dr. P. E. Brown (vol. 31, p. 511).

4. Zeophyllite: by Dr. R. A. Chalmers, Miss L. S. Dent, and Dr. H. F. W. Taylor (vol. 31, p. 726).

5. The powder patterns and lattice parameters of plagioclase felspars. II: by Dr. J. V. Smith and Dr. P. Gay (vol. 31, p. 744).

The following paper was taken as read:

1. Rhodesite, a new mineral from Bultfontein mine, Kimberley, South Africa: by Prof. E. D. Mountain (vol. 31, p. 607).

EXCERPT MINUTES OF COUNCIL MEETING

Held in the Apartments of the Geological Society, Burlington House, London, W., 6 *June* 1957, *at* 3.15 *p.m.*

Prof. L. Hawkes, President, in the Chair.

The following were provisionally elected Ordinary Members of the Society: Arthur Benson Cummins, Pow-Foong Fan, Claude Jean Guy Guillemin, Donald Richard Lewis, Karel Paděra, Irene Marianne Michaelis de Saenz, Julius Schlocker.

The death of Dr. Samuel James Shand was reported.

The reinstatement of Prof. S. B. McCaleb was agreed.

Reports were received from the Treasurer and the Editors, the Editorial Committee on Mineralogical Abstracts, and the Clay Minerals Group.

The Council agreed to the increased subscription rate of 15*s.* for the Clay Minerals Bulletin, and the charge of 10*s.* for single copies and back numbers, commencing 1958.

The General Secretary was nominated by the Council as delegate to a meeting to be held in Montreal in July, to consider the formation of an International Mineralogical Association.

The Council approved the presentation of vols. 30 and 31 (to end of 1957) of the Mineralogical Magazine to the Hungarian National Museum to replenish their destroyed library.

The appointment of Miss A. S. Rotherham as Publications Clerk was reported.

GENERAL MEETING

Held in the Apartments of the Geological Society, Burlington House, London, W., 6 *June* 1957, *at* 5 *p.m.*

Prof. L. Hawkes, President, in the Chair.

Thirty-six Members and four Visitors were present.

The elections of seven Ordinary Members of the Society provisionally made by the Council were confirmed.

The following exhibit was shown:

Ballotini for viewing interference figures of small crystals: by Dr. A. T. J. Dollar.

The following papers were read and discussed:

1. Götzenite and combeite, two new silicates from the Belgian Congo: by Prof. Th. G. Sahama and Mr. Kai Hytönen (vol. 31, p. 503).

2. Isokite and triplite from Bohemia: by Dr. D. Jerome Fisher (vol. 31, p. 587).

3. Heating experiments on some natural titaniferous magnetites: by Dr. E. A. Vincent, Mr. J. B. Wright, Prof. R. Chevallier, and Mlle Suzanne Mathieu (vol. 31, p. 624).

4. An extinction curve method for the universal stage: by Mr. N. Joel and Dr. I. D. Muir (vol. 31, pp. 860 and 878).

The following paper was taken as read:

Measurement and distribution of zircons in some granitic rocks of magmatic origin: by Dr. L. H. Larsen and Dr. A. Poldevaart (vol. 31, p. 544).

EXCERPT MINUTES OF COUNCIL MEETING

Held in the Apartments of the Geological Society, Burlington House, London, W., 7 November 1957, at 3 p.m.

Prof. L. Hawkes, President, in the Chair.

The following were provisionally elected Ordinary Members of the Society: Joyce Marjorie Bond, Bruno Ernst Joseph Brehler, Kevin Charles Burke, Barry Conrad Milne Butler, Charles Gardner Dodd, Wilfred Allan Elders, Charles Henry Emeleus, Katharine Selden Kniskern Mather, Mosobalaje Olaloye Oyawoye, Guy Perrault, Frank Steeley Smith.

The death of Prof. W. N. Benson, Honorary Member, was reported.

The resignations of Mr. H. G. Dines, Mr. M. D. S. Lewis, Mr. G. S. Mockler, Mr. G. M. Stockley, and Prof. L. Weber were accepted.

Reports were received from the Treasurer and the Editors, the Clay Minerals Group, and the Editorial Committee on Mineralogical Abstracts.

The Council expressed appreciation of the Secretary's successful efforts in promoting agreement between the British and American societies regarding the future joint production of Mineralogical Abstracts.

A report was received from the Secretary of his attendance at a

meeting of the (Provisional) International Mineralogical Association i Montreal, in July.

The resignation of Miss A. S. Rotherham as Publications Clerk wa reported.

Dr. W. Campbell Smith was nominated to serve on the Geologica Committee of The Nature Conservancy, the first meeting of which woul be held on 14 November 1957.

Dr. M. K. Wells and Mr. R. K. Harrison were appointed auditors o the Society's accounts for the year ending 31 December 1957.

The Secretary read the draft of the Annual Report of the Council fo the past year, and, with minor alterations, this was approved fo presentation at the ensuing Anniversary Meeting.

GENERAL (ANNIVERSARY) MEETING

Held in the Apartments of the Geological Society, Burlington House, London, W., 7 November 1957, at 5 p.m.

Prof. L. Hawkes, President, in the Chair.

Forty-eight Members and five Visitors were present.

The elections of eleven Ordinary Members of the Society provisionall made by the Council were confirmed.

The General Secretary read the following:

ANNUAL REPORT OF THE COUNCIL FOR 1956–7

Membership.—The total membership on 31 October 1957 was 637 against 629 o 31 October 1956. Thirty-four new members have been elected during the year nine members have resigned, and the names of five members have been removed o account of arrears of subscription. The Society has lost by death ten Ordinary an two Honorary Members. On 31 October 1957 the membership consisted of fiv Honorary Members and 632 Ordinary Members, of whom 65 are Life Members 287 members are also members of the Clay Minerals Group.

Meetings.—The total attendance at the four general meetings held during th session 1956–7 was 157 members and 20 guests, giving an average of 44 per meeting. At these meetings 18 papers have been read; 11 have been taken as read, and exhibits have been shown.

At two meetings of the Clay Minerals Group, held in London and Oxford re spectively, 18 papers were read: the average attendance was 40 per meeting.

Publications.—Three numbers of the Mineralogical Magazine have been issue during the year, and the fourth (September 1957) is expected shortly. The fou numbers 235–8 contain Proceedings (15 pages), 32 papers, and 17 short communica tions in 324 pages, with 8 plates and 85 text-figures; and 1,150 abstracts, 64 book reviews in 328 pages.

Three numbers (15–17) of the Clay Minerals Bulletin have been issued during the

year. The annual subscription for two issues will be raised to 15*s.* from 1 January 1958.

A new monograph on clay minerals was published in March, entitled, 'The differential thermal investigation of clays', and edited by R. C. Mackenzie.

Financial.—The accounts for the calendar year 1956 are published in the June (1957) number of the Magazine. The excess of assets (exclusive of the Society's library and stock of publications) over liabilities at 31 December 1956 was £7,635. 15*s.* against £7,429. 9*s.* 9*d.* at the close of 1955.

During 1956 income from the General Fund exceeded expenditure by £71. 17*s.* In this, the first complete year of the Society's occupation of its new premises, general office expenses, including clerical assistance, amounted to £1,142. 1*s.* 1*d.*

The two numbers of the Mineralogical Magazine issued since the beginning of 1957 have cost £1,406. 3*s.* 9*d.* (inclusive of authors' reprints). The increased cost over the corresponding two numbers for 1956 is partly due to an increase of size (351 pages against 318), but printing costs have increased by approximately 10 per cent. Sales of the Clay Minerals Group monograph on differential thermal analysis have amounted to £657. 7*s.* 7*d.* Since the last Annual Report was presented grants to the value of £300 have been received from the Royal Society (Parliamentary Grant-in-aid for Scientific Publications) towards the cost of publication of the Magazine and Abstracts and £400 towards the cost of production of a revised edition of the monograph on X-ray identification and crystal structures of clay minerals.

The balances at the Society's Bankers and in the Post Office Savings Bank on 31 October 1957 were: General Fund £3,545. 10*s.* 9*d.*, Miers Memorial Fund £125. 10*s.* 2*d.*

General.—i. *Mineralogical Abstracts*: Negotiations have been proceeding during the year with the Mineralogical Society of America with the object of increasing the size and coverage of Mineralogical Abstracts. It is hoped that an arrangement will be concluded for the joint production of the Abstracts by the two Societies from the commencement of 1959. A full report will be presented to the Society when arrangements have been completed.

ii. *International Mineralogical Association*: A meeting of delegates of Mineralogical Societies of national status was held in Montreal during the International Crystallographical Congress in July 1957 to consider the formation of the International Mineralogical Association. It was agreed that such an Association was desirable; a meeting for its formal inauguration was arranged for Madrid in the Spring of 1958.

On the proposal of Dr. A. F. Hallimond, seconded by Mr. G. E. Howling, the adoption of the Annual Report was carried unanimously.

A vote of thanks to the Geological Society for the use of the apartments during the past session was proposed by Prof. K. C. Dunham, seconded by Dr. A. T. J. Dollar, and carried with acclamation.

The President read the following list of Officers and Council elected for the session 1957–8 in accordance with by-law 20:

President. Prof. C. E. Tilley, B.Sc., Ph.D., F.R.S.

Vice-Presidents. Prof. L. R. Wager, M.A., ScD., B.Sc., F.R.S.; Dr. A. K. Wells, D.Sc., F.G.S.

[*continued on p. lxxx*

MINERALOGIC

INCOME AND EXPENDITURE ACCOU

A. GENER

1956 £	Income	£ s. d.	£
	Members' subscriptions—		
	Annual subscriptions	1139 1 3	
	Entrance fees	33 0 0	
1138			1172
	Mineralogical Magazine and Abstracts—		
	Advertisement fees	79 3 6	
	Sales	3896 18 6	
2123			3976
	Investment revenue—		
	Interest on investments	207 14 0	
	Post Office Savings Bank interest	56 16 10	
256			264 1
550	Grant from Royal Society (Parliamentary Grant-in-aid for scientific publications)		300
	Clay Minerals Group—		
	Clay Minerals Bulletin:		
	Subscriptions	94 13 8	
	Sales, including authors' reprints	201 5 7	
248			295 1
	Clay Minerals Monographs—		
	Sales: *X-ray Identification and Crystal Structures of Clay Minerals*	5 15 4	
–	*The Differential Thermal Investigation of Clays*	763 14 5	
			769
16	*Glossary of Clay Trade Names*, sales		12
–	Interest on Bank Account (Cambridge)		1 1
–	Transfer from Publications Fund		1000
–	Balance, being excess of expenditure over income for the year		175
£4338			£7967

B. MI

£		£
18	Investment revenue	17 1
1	Sundry revenue	1
£19		£18 1

OCIETY

R THE YEAR ENDED 31 DECEMBER 1957

ND

£	Expenditure	£ s. d.	£ s. d.
	Mineralogical Magazine and Abstracts—		
	Nos. 236–9:		
	Printing	2561 11 10	
	Illustrations	163 18 4	
	Authors' reprints	172 0 3	
	Distribution to members	86 1 7	
	„ „ subscribers	111 3 6	
04			3094 15 6
78	Reprinting Magazines Nos. 169, 170, 174, 179, 192		565 12 9
	Editorial expenses—		
	Magazine editor	57 18 5	
	Abstracts editor	230 17 9	
32			288 16 2
	Meeting expenses—		
	Notices of meetings	86 6 8	
	General expenses in connexion with meetings	26 8 0	
10			112 14 8
	General expenses—		
	Office rental and house expenses	404 16 11	
	Clerical assistance	991 1 6	
	Printing and stationery	94 3 2	
	Postage and telephone	123 8 10	
	Sundry office expenses	30 16 2	
42			1644 6 7
28	*Miscellaneous charges*		– – –
	Clay Minerals Group—		
	Clay Minerals Bulletin:		
	Printing Bulletins 16 and 17	200 16 6	
	Authors' reprints Bulletin 15	27 10 0	
	Editorial expenses	15 10 11	
03			243 17 5
	Monograph on *The Differential Thermal Investigation of Clays*:		
	Printing and binding Monograph	1917 0 0	
—	Cardboard cartons for distribution of Monograph	24 10 7	
			1941 10 7
	General expenses:		
	Meeting expenses	19 6 0	
	Office expenses	56 6 11	
69			75 12 11
72	*Balance, being excess of income over expenditure for the year*		– – –
38			£7967 6 7

EMORIAL FUND

£		£ s. d.
–	Bank charges	2 8
19	*Balance*, being excess of income over expenditure for the year	18 11 6
19		18 14 2

James H. Taylor, Hon. Treasurer.

M. K. Wells
R. K. Harrison } Auditors

March 1958.

MINERALOGIC

BALANCE SHEET AS

A. GENE

1956 £	Liabilities	£ s. d.	£ s. d.	£
	Fund Account—			
	Balance per Balance Sheet at 31 December 1956	7080 14 1		
	Add Composition Fees received	119 18 4		
			7200 12 5	
	Less Depreciation on office furniture and equipment	57 10 9		
	Excess of expenditure over income for year ended 31 December 1957	175 4 2		
			232 14 11	
7081				6967
2478	Publications Fund			1477
	Subscriptions, &c., received in advance—			
	Members' subscriptions and entrance fees		92 19 5	
	Subscriptions to *Clay Minerals Bulletin*		2 0 0	
94				94
	Sundry creditors—			
	Oxford University Press for Magazine No. 239		677 11 6	
	Royal Entomological Society for house expenses		39 15 6	
	Editors for sundry editorial expenses		95 3 0	
676				812
£10329				£9353

B. MI

£		£ s. d.	£
	Fund Account—		
	Balance per Balance Sheet at 31 December 1956	555 0 11	
	Add Excess of income over expenditure for the year ended 31 December 1957	18 11 6	
555			573
£555			£573

OCIETY

DECEMBER 1957

ND

956 £	ASSETS	£ s. d.	£ s. d.
	Office furniture and equipment—		
	Original cost	575 7 5	
	Less depreciation	211 10 8	
421			363 16 9
	Investments (at cost or as valued at 31 December 1951)—		
	£2232. 9*s.* 3½% War Stock 1952	1856 8 6	
	£753. 12*s.* 11*d.* 4% Consolidated Stock 1957	694 4 9	
	£606. 4*s.* 6*d.* 4% Funding Stock 1960–90	598 5 0	
	£250 3% Savings Bonds 1960–70	216 17 6	
	£1013. 9*s.* 6*d.* 3% Savings Bonds 1965–75	916 7 0	
	£100 3% British Transport Stock 1978–88	77 0 0	
	£275 2½% Consolidated Stock	169 2 6	
	£261. 12*s.* 6*d.* 3½% Treasury Stock 1977–80	244 8 3	
	£850 4½% British Electricity Stock 1967–69	810 12 5	
537			5583 5 11
	Note:—The total market value of the above investments at 31 December 1957 was £4518. 14*s.* 8*d.*		
	Deposits—		
857	Balance with Post Office Savings Bank		1913 17 10
	Subscriptions, &c., in arrears—		
	Members' subscriptions	44 8 1	
106	Subscriptions to *Clay Minerals Bulletin*	6 0 0	
	Sundry debtors—		50 8 1
	Authors for reprints from *Clay Minerals Bulletin*	14 10 0	
	Purchasers of *Clay Minerals Bulletin*	28 15 0	
986			43 5 0
	Cash—		
	Coutts & Co., London	838 5 2	
	Barclays Bank, Harpenden	452 4 10	
	Cambridge & District Trustee Savings Bank	77 14 2	
	Cash in hand, London	20 0 0	
	" " Cambridge (Abstracts Editor)	10 0 0	
	" " Harpenden	2 11	
422			1398 7 1
329			£9353 0 8

Note:—The value of the Society's library and stock of publications is not included in the above assets.

EMORIAL FUND

£		£ s. d.
	Investments (at cost)—	
439	£504. 17*s.* 4*d.* 3½% War Stock 1952	439 7 0
	Note:—The total market value of the above investment at 31 December 1957 was £318. 1*s.* 3*d.*	
116	Cash—Coutts & Co., London	134 5 5
555		£573 12 5

JAMES H. TAYLOR, Hon. Treasurer.

M. K. WELLS
R. K. HARRISON } Auditors

13 *March* 1958

Treasurer. Prof. J. H. Taylor, M.A., B.Sc., Ph.D., F.G.S.
General Secretary. Dr. G. F. Claringbull, B.Sc., Ph.D., A.Inst.P., F.G.S.
Foreign Secretary. Dr. L. J. Spencer, C.B.E., B.Sc., M.A., Sc.D., F.R.S.
Editor of the Journal. Dr. M. H. Hey, M.A., D.Sc.
Editor of Mineralogical Abstracts. Dr. N. F. M. Henry, B.Sc., M.A., Ph.D., F.G.S.

Ordinary Members of Council

Dr. G. M. Brown, M.A., D.Phil., B.Sc., F.G.S.
Prof. W. A. Deer, M.Sc., Ph.D., F.G.S.
Dr. H. I. Drever, B.Sc., Ph.D., F.R.S.E.
Mr. P. G. Embrey, M.A., B.Sc., A.R.I.C.
Mr. J. E. T. Horne, M.A., F.G.S.
Dr. R. C. Mackenzie, B.Sc., Ph.D., A.R.I.C.
Dr. R. J. W. McLaughlin, M.Sc., Ph.D.
Dr. A. F. Seager, B.Sc., Ph.D., A.K.C., F.G.S.
Prof. A. Stuart, M.Sc., F.G.S., F.R.M.S.
Dr. W. H. Taylor, M.A., Ph.D., D.Sc., F.Inst.P.
Dr. E. A. Vincent, M.A., B.Sc., Ph.D., F.R.I.C., F.G.S.
Prof. D. Williams, D.Sc., Ph.D., B.E., M.I.M.M., F.G.S.

The retiring President, Prof. L. Hawkes, in welcoming Prof. Tilley to the Chair, thanked the Officers of the Society for their help during his period of office, and expressed the hope that Prof. Tilley's presidency would be a happy one.

Prof. Tilley, in reply, thanked the Society for his election and expressed appreciation of Prof. Hawkes's guidance of the Society during the past three years. He then proposed a vote of thanks to the retiring President and Ordinary Members of the Council which was carried with acclamation.

The following exhibits were shown:

1. Some minerals new to Britain: by Mr. A. W. G. Kingsbury.

2. A high-temperature microscope furnace for petrological analysis employing a thermocouple as heater and thermometer combined: by Dr. A. T. J. Dollar and Mr. B. C. Hicks.

The following papers were read:

1. Cornubite ($Cu_5(AsO_4)_2(OH)_4$), a new mineral from Cornwall: by Dr. G. F. Claringbull, Dr. M. H. Hey, and Sir Arthur Russell.

2. Rare earths in some niobate-tantalates: by Dr. J. R. Butler (vol. 31, p. 763).

3. Plagioclases from the Juvinas meteorite and from allivalite from the Isle of Rhum: by Mr. P. M. Game (vol. 31, p. 656).

4. The alteration of pyrochlore to columbite in carbonatites in Tanganyika: by Mr. T. C. James and Mr. Duncan McKie.

5. Third supplementary list of British minerals: by Dr. L. J. Spencer (vol. 31, p. 787).

6. On the unit cell of synthetic and natural syngenite: by Dr. E. Aruja.

The following papers were taken as read:

1. Some zoned skarns from granite-marble contacts near Puyvalador, in the Quérigut area, eastern Pyrenees, and their petrogenesis: by Dr. W. A. Watters (vol. 31, p. 703).

2. Sapphire-bearing rocks from Antarctica: by Dr. E. R. Segnit (vol. 31, p. 690).